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(54) Abstract Title

**Stabilizer mixture for organic materials**

(57) Organic materials which possess outstanding stability to oxidative, thermal or light-induced degradation comprise as stabilizers at least one compound of the benzofuran-2-one type, at least one compound of the organic phosphite or phosphonite type, at least one compound of the phenolic antioxidant type and at least one compound of the sterically hindered amine type.

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Stabilizer mixture for organic materials

The present invention relates to compositions comprising an organic material subject to oxidative, thermal or light-induced degradation, and as stabilizers at least one compound of the benzofuran-2-one type, at least one compound of the phosphite or phosphonite type, at least one compound of the phenolic antioxidant type and at least one compound of the sterically hindered amine type, and to the use thereof for stabilizing organic materials against oxidative, thermal or light-induced degradation.

The use of compounds of the benzofuran-2-one type as stabilizers for organic polymers is known, for example, from US 4,325,863; US 4,388,244; US 5,175,312; US 5,252,643; US 5,216,052; US 5,369,159; US 5,488,117; US 5,356,966; US 5,367,008; US 5,428,162; US 5,428,177 or US 5,516,920.

Organic phosphites, phosphonites and phosphoramides are known in the art as costabilizers, secondary antioxidants and processing stabilizers, inter alia for polyolefins. Examples of such known phosphite stabilizers can be found in R. Gächter/H. Müller (eds.), *Plastics Additives Handbook*, 3rd ed., page 47, Hanser, Munich 1990.

U.S. Patent 4,360,617 discloses that stabilizer mixtures comprising symmetrical triarylphosphites and certain phenolic antioxidants are particularly suitable for protecting certain organic materials, such as polyurethane, polyacrylonitrile, polyamide 12 or polystyrene, against oxidative, thermal or light-induced degradation.

Sterically hindered amines, including in particular compounds containing 2,2,6,6-tetramethylpiperidyl groups, are known as hindered amine light stabilizers (HALS).

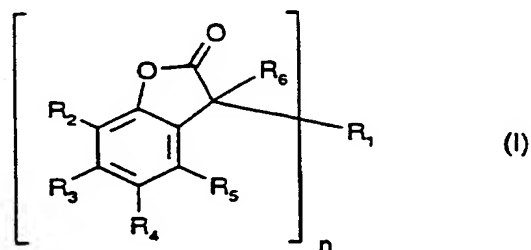
The known stabilizers do not satisfy in every respect the high requirements which a stabilizer is required to meet, especially with regard to shelf life, water absorption, sensitivity to hydrolysis, in-process stabilization, colour properties, volatility, migration behaviour, compatibility and improvement in protection against light. As a result there continues to be a need for effective stabilizers for organic materials that are sensitive to oxidative, thermal and/or light-induced degradation.

It has now been found that a stabilizer mixture comprising at least one compound of the benzofuran-2-one type, at least one compound of the organic phosphite or phosphonite type, at least one compound of the phenolic antioxidant type and at least one compound of the sterically hindered amine type is particularly suitable as a stabilizer for organic materials which are sensitive to oxidative, thermal or light-induced degradation.

The present invention therefore provides compositions comprising

- a) an organic material which is subject to oxidative, thermal or light-induced degradation,
- b) at least one compound of the benzofuran-2-one type,
- c) at least one compound from the group of the organic phosphites or phosphonites,
- d) at least one compound from the group of the phenolic antioxidants, and
- e) at least one compound from the group of the sterically hindered amines.

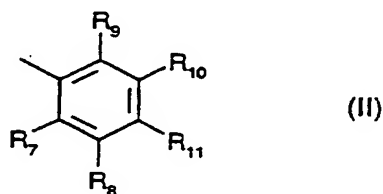
Compositions which are of interest include those comprising as component (b) a compound of the formula I



in which, if n is 1,

R<sub>1</sub> is unsubstituted or C<sub>1</sub>-C<sub>4</sub>alkyl-, C<sub>1</sub>-C<sub>4</sub>alkoxy-, C<sub>1</sub>-C<sub>4</sub>alkylthio-, hydroxyl-, halo-, amino-, C<sub>1</sub>-C<sub>4</sub>alkylamino-, phenylamino- or di(C<sub>1</sub>-C<sub>4</sub>alkyl)amino-substituted naphthyl, phenanthryl, anthryl, 5,6,7,8-tetrahydro-2-naphthyl, 5,6,7,8-tetrahydro-1-naphthyl, thienyl, benzo[b]thienyl, naphtho[2,3-b]thienyl, thianthrenyl, dibenzofuryl, chromenyl, xanthenyl, phenoxathiinyl, pyrrolyl, imidazolyl, pyrazolyl, pyrazinyl, pyrimidinyl, pyridazinyl, indoliziny, isoindolyl, indolyl, indazolyl, purinyl, quinoliziny, isoquinolyl, quinolyl, phthalazinyl, naphthyridinyl, quinoxalinyl, quinazolinyl, cinnolinyl, pteridinyl, carbazolyl, β-carbolinyl, phenanthridinyl, acridinyl,

perimidinyl, phenanthrolinyl, phenazinyl, isothiazolyl, phenothiazinyl, isoxazolyl, furazanyl, biphenyl, terphenyl, fluorenyl or phenoxazinyl, or  $R_1$  is a radical of the formula II



and

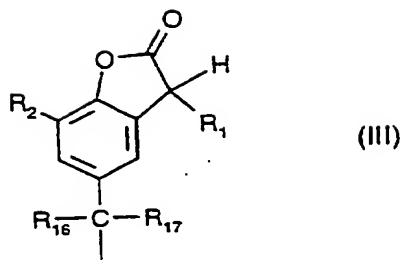
if  $n$  is 2,

$R_1$  is unsubstituted or  $C_1$ - $C_4$ alkyl- or hydroxyl-substituted phenylene or naphthylene; or is  $-R_{12}-X-R_{13}-$ ,

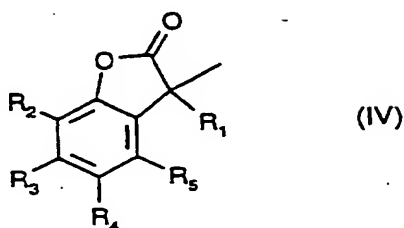
$R_2$ ,  $R_3$ ,  $R_4$  and  $R_5$  independently of one another are hydrogen, chlorine, hydroxyl,  $C_1$ - $C_{25}$ alkyl,  $C_7$ - $C_9$ phenylalkyl, unsubstituted or  $C_1$ - $C_4$ alkyl-substituted phenyl; unsubstituted or  $C_1$ - $C_4$ alkyl-substituted  $C_5$ - $C_8$ cycloalkyl;  $C_1$ - $C_{18}$ alkoxy,  $C_1$ - $C_{18}$ alkylthio,  $C_1$ - $C_4$ alkylamino, di( $C_1$ - $C_4$ alkyl)amino,  $C_1$ - $C_{25}$ alkanoyloxy,  $C_1$ - $C_{25}$ alkanoylamino,  $C_3$ - $C_{25}$ alkenoyloxy,

$C_3$ - $C_{25}$ alkanoyloxy which is interrupted by oxygen, sulfur or  $\text{>N}-R_{14}$  ;  $C_6$ - $C_9$ cycloalkyl-

carbonyloxy, benzoyloxy or  $C_1$ - $C_{12}$ alkyl-substituted benzoyloxy; or else the radicals  $R_2$  and  $R_3$  or the radicals  $R_3$  and  $R_4$  or the radicals  $R_4$  and  $R_5$ , together with the carbon atoms to which they are attached, form a benzo ring,  $R_4$  is additionally  $-(CH_2)_p-COR_{15}$  or  $-(CH_2)_qOH$  or, if  $R_3$ ,  $R_5$  and  $R_6$  are hydrogen,  $R_4$  is additionally a radical of the formula III



in which  $R_1$  is defined as indicated above for  $n = 1$ ,  
 $R_6$  is hydrogen or a radical of the formula IV



where  $R_4$  is not a radical of the formula III and  $R_1$  is defined as indicated above for  $n = 1$ ,  
 $R_7, R_8, R_9, R_{10}$  and  $R_{11}$  independently of one another are hydrogen, halogen, hydroxyl,

$C_1$ - $C_{25}$ alkyl,  $C_2$ - $C_{25}$ alkyl interrupted by oxygen, sulfur or  $\text{>N-R}_{14}$  ;  $C_1$ - $C_{25}$ alkoxy,

$C_2$ - $C_{25}$ alkoxy interrupted by oxygen, sulfur or  $\text{>N-R}_{14}$  ;  $C_1$ - $C_{25}$ alkylthio,  $C_3$ - $C_{25}$ alkenyl,

$C_3$ - $C_{25}$ alkenyloxy,  $C_3$ - $C_{25}$ alkynyl,  $C_3$ - $C_{25}$ alkynyloxy,  $C_7$ - $C_9$ phenylalkyl,  $C_7$ - $C_9$ phenylalkoxy,  
 unsubstituted or  $C_1$ - $C_4$ alkyl-substituted phenyl; unsubstituted or  $C_1$ - $C_4$ alkyl-substituted  
 phenoxy; unsubstituted or  $C_1$ - $C_4$ alkyl-substituted  $C_5$ - $C_8$ cycloalkyl; unsubstituted or  
 $C_1$ - $C_4$ alkyl-substituted  $C_5$ - $C_8$ cycloalkoxy;  $C_1$ - $C_4$ alkylamino, di( $C_1$ - $C_4$ alkyl)amino,

$C_1$ - $C_{25}$ alkanoyl,  $C_3$ - $C_{25}$ alkanoyl interrupted by oxygen, sulfur or  $\text{>N-R}_{14}$  ;

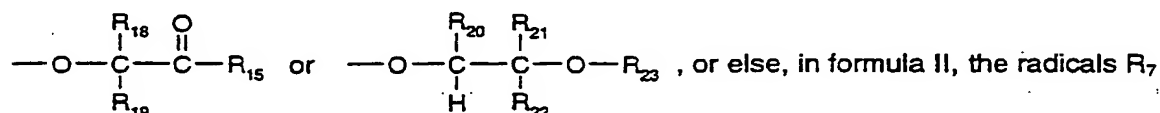
$C_1$ - $C_{25}$ alkanoyloxy,  $C_3$ - $C_{25}$ alkanoyloxy interrupted by oxygen, sulfur or  $\text{>N-R}_{14}$  ;

C<sub>1</sub>-C<sub>25</sub>alkanoylamino, C<sub>3</sub>-C<sub>25</sub>alkenoyl, C<sub>3</sub>-C<sub>25</sub>alkenoyl interrupted by oxygen, sulfur or

$\text{>N-R}_{14}$  ; C<sub>3</sub>-C<sub>25</sub>alkenoyloxy, C<sub>3</sub>-C<sub>25</sub>alkenoyloxy interrupted by oxygen, sulfur or

$\text{>N-R}_{14}$  ; C<sub>6</sub>-C<sub>9</sub>cycloalkylcarbonyl, C<sub>6</sub>-C<sub>9</sub>cycloalkylcarbonyloxy, benzoyl or C<sub>1</sub>-C<sub>12</sub>alkyl-

substituted benzoyl; benzoyloxy or C<sub>1</sub>-C<sub>12</sub>alkyl-substituted benzoyloxy;



and R<sub>8</sub> or the radicals R<sub>8</sub> and R<sub>11</sub>, together with the carbon atoms to which they are attached, form a benzo ring,

R<sub>12</sub> and R<sub>13</sub> independently of one another are unsubstituted or C<sub>1</sub>-C<sub>4</sub>alkyl-substituted phenylene or naphthylene,

R<sub>14</sub> is hydrogen or C<sub>1</sub>-C<sub>8</sub>alkyl,

R<sub>15</sub> is hydroxyl,  $\left[ -\text{O}^- \frac{1}{r} \text{M}^{r+} \right]$ , C<sub>1</sub>-C<sub>18</sub>alkoxy or  $\text{>N} \begin{array}{l} \text{R}_{24} \\ \text{R}_{25} \end{array}$ ,

R<sub>16</sub> and R<sub>17</sub> independently of one another are hydrogen, CF<sub>3</sub>, C<sub>1</sub>-C<sub>12</sub>alkyl or phenyl, or R<sub>16</sub> and R<sub>17</sub>, together with the C atom to which they are attached, form a C<sub>5</sub>-C<sub>8</sub>cycloalkylidene ring which is unsubstituted or substituted from 1 to 3 times by C<sub>1</sub>-C<sub>4</sub>alkyl;

R<sub>18</sub> and R<sub>19</sub> independently of one another are hydrogen, C<sub>1</sub>-C<sub>4</sub>alkyl or phenyl,

R<sub>20</sub> is hydrogen or C<sub>1</sub>-C<sub>4</sub>alkyl,

R<sub>21</sub> is hydrogen, unsubstituted or C<sub>1</sub>-C<sub>4</sub>alkyl-substituted phenyl; C<sub>1</sub>-C<sub>25</sub>alkyl, C<sub>2</sub>-C<sub>25</sub>alkyl

interrupted by oxygen, sulfur or  $\text{>N-R}_{14}$  ; C<sub>7</sub>-C<sub>9</sub>phenylalkyl which is unsubstituted or

substituted on the phenyl radical from 1 to 3 times by C<sub>1</sub>-C<sub>4</sub>alkyl; C<sub>7</sub>-C<sub>25</sub>phenylalkyl which is unsubstituted or substituted on the phenyl radical from 1 to 3 times by C<sub>1</sub>-C<sub>4</sub>alkyl and

interrupted by oxygen, sulfur or  $\text{>N-R}_{14}$  , or else the radicals R<sub>20</sub> and R<sub>21</sub>, together with

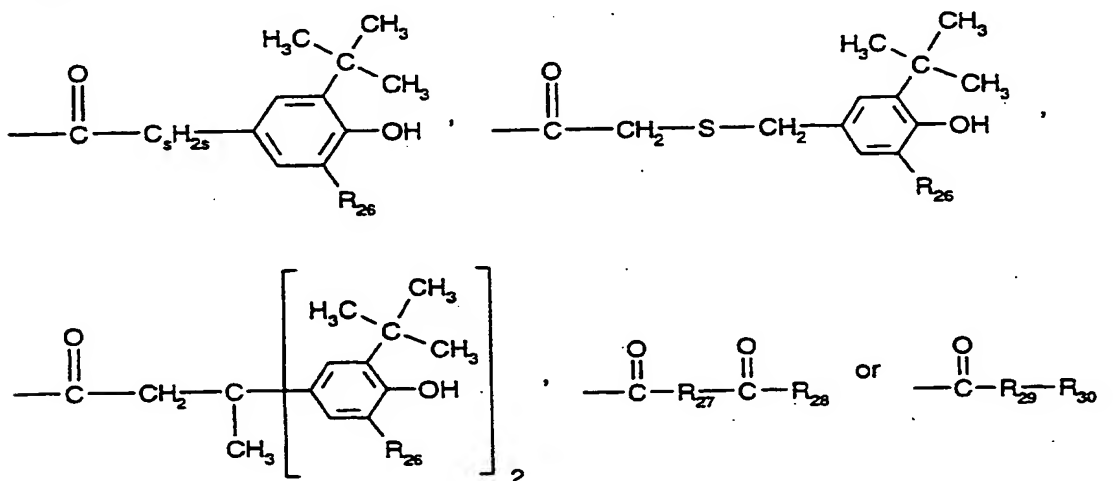
the carbon atoms to which they are attached, form a C<sub>5</sub>-C<sub>12</sub>cycloalkylene ring which is unsubstituted or substituted from 1 to 3 times by C<sub>1</sub>-C<sub>4</sub>alkyl;

$R_{22}$  is hydrogen or  $C_1$ - $C_4$ alkyl,

$R_{23}$  is hydrogen,  $C_1$ - $C_{25}$ alkanoyl,  $C_3$ - $C_{25}$ alkenoyl,  $C_3$ - $C_{25}$ alkanoyl interrupted by oxygen, sulfur

or  $\text{>N}-R_{14}$  ;  $C_2$ - $C_{25}$ alkanoyl substituted by a di( $C_1$ - $C_6$ alkyl)phosphonate group;

$C_6$ - $C_9$ cycloalkylcarbonyl, thenoyl, furoyl, benzoyl or  $C_1$ - $C_{12}$ alkyl-substituted benzoyl;



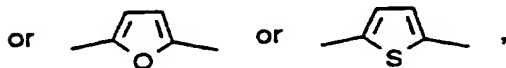
$R_{24}$  and  $R_{25}$  independently of one another are hydrogen or  $C_1$ - $C_{18}$ alkyl,

$R_{26}$  is hydrogen or  $C_1$ - $C_8$ alkyl,

$R_{27}$  is a direct bond,  $C_1$ - $C_{18}$ alkylene,  $C_2$ - $C_{18}$ alkylene interrupted by oxygen, sulfur or

$\text{>N}-R_{14}$  ;  $C_2$ - $C_{18}$ alkenylene,  $C_2$ - $C_{20}$ alkylidene,  $C_7$ - $C_{20}$ phenylalkylidene,

$C_5$ - $C_8$ cycloalkylene,  $C_7$ - $C_8$ bicycloalkylene, unsubstituted or  $C_1$ - $C_4$ alkyl-substituted phenylene,



$R_{28}$  is hydroxyl,  $\left[ -O^- \frac{1}{r} M^{r+} \right]$ ,  $C_1$ - $C_{18}$ -alkoxy or  $\text{>N}-R_{24}$  ,

$R_{29}$  is oxygen, -NH- or  $\text{>N}-C(=O)-NH-R_{30}$  ,

$R_{30}$  is  $C_1$ - $C_{18}$ alkyl or phenyl,



R<sub>31</sub> is hydrogen or C<sub>1</sub>-C<sub>18</sub>alkyl,

M is an r-valent metal cation,

X is a direct bond, oxygen, sulfur or -NR<sub>31</sub>-,

n is 1 or 2,

p is 0, 1 or 2,

q is 1, 2, 3, 4, 5 or 6,

r is 1, 2 or 3, and

s is 0, 1 or 2.

Unsubstituted or C<sub>1</sub>-C<sub>4</sub>alkyl-, C<sub>1</sub>-C<sub>4</sub>alkoxy-, C<sub>1</sub>-C<sub>4</sub>alkylthio-, hydroxyl-, halo-, amino-, C<sub>1</sub>-C<sub>4</sub>alkylamino-, phenylamino- or di(C<sub>1</sub>-C<sub>4</sub>alkyl)amino-substituted naphthyl, phenanthryl, anthryl, 5,6,7,8-tetrahydro-2-naphthyl, 5,6,7,8-tetrahydro-1-naphthyl, thienyl, benzo[b]thienyl, naphtho[2,3-b]thienyl, thianthrenyl, dibenzofuryl, chromenyl, xanthenyl, phenoxathiinyl, pyrrolyl, imidazolyl, pyrazolyl, pyrazinyl, pyrimidinyl, pyridazinyl, indoliziny, isoindolyl, indolyl, indazolyl, purinyl, quinoliziny, isoquinolyl, quinolyl, phthalazinyl, naphthyridinyl, quinoxalinyl, quinazolinyl, cinnoliny, pteridinyl, carbazolyl,  $\beta$ -carboliny, phenanthridinyl, acridinyl, perimidinyl, phenanthrolinyl, phenazinyl, isothiazolyl, phenothiazinyl, isoxazolyl, furazanyl, biphenyl, terphenyl, fluorenyl or phenoxazinyl is, for example, 1-naphthyl, 2-naphthyl, 1-phenylamino-4-naphthyl, 1-methylnaphthyl, 2-methylnaphthyl, 1-methoxy-2-naphthyl, 2-methoxy-1-naphthyl, 1-dimethylamino-2-naphthyl, 1,2-dimethyl-4-naphthyl, 1,2-dimethyl-6-naphthyl, 1,2-dimethyl-7-naphthyl, 1,3-dimethyl-6-naphthyl, 1,4-dimethyl-6-naphthyl, 1,5-dimethyl-2-naphthyl, 1,6-dimethyl-2-naphthyl, 1-hydroxy-2-naphthyl, 2-hydroxy-1-naphthyl, 1,4-dihydroxy-2-naphthyl, 7-phenanthryl, 1-anthryl, 2-anthryl, 9-anthryl, 3-benzo[b]thienyl, 5-benzo[b]thienyl, 2-benzo[b]thienyl, 4-dibenzofuryl, 4,7-dibenzofuryl, 4-methyl-7-dibenzofuryl, 2-xanthenyl, 8-methyl-2-xanthenyl, 3-xanthenyl, 2-phenoxathiinyl, 2,7-phenoxathiinyl, 2-pyrrolyl, 3-pyrrolyl, 5-methyl-3-pyrrolyl, 2-imidazolyl, 4-imidazolyl, 5-imidazolyl, 2-methyl-4-imidazolyl, 2-ethyl-4-imidazolyl, 2-ethyl-5-imidazolyl, 3-pyrazolyl, 1-methyl-3-pyrazolyl, 1-propyl-4-pyrazolyl, 2-pyrazinyl, 5,6-dimethyl-2-pyrazinyl, 2-indoliziny, 2-methyl-3-isoindolyl, 2-methyl-1-isoindolyl, 1-methyl-2-indolyl, 1-methyl-3-indolyl, 1,5-dimethyl-2-indolyl, 1-methyl-3-indazolyl, 2,7-dimethyl-8-purinyl, 2-methoxy-7-methyl-8-purinyl, 2-quinoliziny, 3-isoquinolyl, 6-isoquinolyl, 7-isoquinolyl, isoquinolyl, 3-methoxy-6-isoquinolyl, 2-quinolyl, 6-quinolyl, 7-quinolyl, 2-methoxy-3-quinolyl, 2-methoxy-6-quinolyl, 6-phthalazinyl, 7-phthalazinyl, 1-methoxy-6-phthalazinyl, 1,4-dimethoxy-6-phthalazinyl, 1,8-naphthyridin-2-yl, 2-quinoxalinyl, 6-quinoxalinyl, 2,3-dimethyl-6-quinoxalinyl, 2,3-dimethoxy-6-quinoxalinyl, 2-quinazolinyl, 7-quinazolinyl,

2-dimethylamino-6-quinazolinyl, 3-cinnolinyl, 6-cinnolinyl, 7-cinnolinyl, 3-methoxy-7-cinnolinyl, 2-pteridiny, 6-pteridiny, 7-pteridiny, 6,7-dimethoxy-2-pteridiny, 2-carbazolyl, 3-carbazolyl, 9-methyl-2-carbazolyl, 9-methyl-3-carbazolyl,  $\beta$ -carbolin-3-yl, 1-methyl- $\beta$ -carbolin-3-yl, 1-methyl- $\beta$ -carbolin-6-yl, 3-phenanthridinyl, 2-acridinyl, 3-acridinyl, 2-perimidinyl, 1-methyl-5-perimidinyl, 5-phenanthrolinyl, 6-phenanthrolinyl, 1-phenazinyl, 2-phenazinyl, 3-isothiazolyl, 4-isothiazolyl, 5-isothiazolyl, 2-phenothiazinyl, 3-phenothiazinyl, 10-methyl-3-phenothiazinyl, 3-isoxazolyl, 4-isoxazolyl, 5-isoxazolyl, 4-methyl-3-furazanyl, 2-phenoxazinyl or 10-methyl-2-phenoxazinyl.

Particular preference is given to unsubstituted or  $C_1$ - $C_4$ alkyl-,  $C_1$ - $C_4$ alkoxy-,  $C_1$ - $C_4$ alkylthio-, hydroxyl-, phenylamino- or di( $C_1$ - $C_4$ alkyl)amino-substituted naphthyl, phenanthryl, anthryl, 5,6,7,8-tetrahydro-2-naphthyl, 5,6,7,8-tetrahydro-1-naphthyl, thienyl, benzo[b]thienyl, naphtho[2,3-b]thienyl, thianthrenyl, dibenzofuryl, chromenyl, xanthenyl, phenoxathiinyl, pyrrolyl, isoindolyl, indolyl, phenothiazinyl, biphenyl, terphenyl, fluorenyl or phenoxazinyl, for example 1-naphthyl, 2-naphthyl, 1-phenylamino-4-naphthyl, 1-methylnaphthyl, 2-methylnaphthyl, 1-methoxy-2-naphthyl, 2-methoxy-1-naphthyl, 1-dimethylamino-2-naphthyl, 1,2-dimethyl-4-naphthyl, 1,2-dimethyl-6-naphthyl, 1,2-dimethyl-7-naphthyl, 1,3-dimethyl-6-naphthyl, 1,4-dimethyl-6-naphthyl, 1,5-dimethyl-2-naphthyl, 1,6-dimethyl-2-naphthyl, 1-hydroxy-2-naphthyl, 2-hydroxy-1-naphthyl, 1,4-dihydroxy-2-naphthyl, 7-phenanthryl, 1-anthryl, 2-anthryl, 9-anthryl, 3-benzo[b]thienyl, 5-benzo[b]thienyl, 2-benzo[b]thienyl, 4-dibenzofuryl, 4,7-dibenzofuryl, 4-methyl-7-dibenzofuryl, 2-xanthenyl, 8-methyl-2-xanthenyl, 3-xanthenyl, 2-pyrrolyl, 3-pyrrolyl, 2-phenothiazinyl, 3-phenothiazinyl, 10-methyl-3-phenothiazinyl.

Halogen (halo) is, for example, chlorine, bromine or iodine. Preference is given to chlorine.

Alkanoyl having up to 25 carbon atoms is a branched or unbranched radical, for example formyl, acetyl, propionyl, butanoyl, pentanoyl, hexanoyl, heptanoyl, octanoyl, nonanoyl, decanoyl, undecanoyl, dodecanoyl, tridecanoyl, tetradecanoyl, pentadecanoyl, hexadecanoyl, heptadecanoyl, octadecanoyl, eicosanoyl or docosanoyl. Preference is given to alkanoyl having 2 to 18, especially 2 to 12, for example 2 to 6 carbon atoms. Particular preference is given to acetyl.

$C_2$ - $C_{25}$ alkanoyl substituted by a di( $C_1$ - $C_6$ alkyl)phosphonate group is, for example,  $(CH_3CH_2O)_2POCH_2CO-$ ,  $(CH_3O)_2POCH_2CO-$ ,  $(CH_3CH_2CH_2CH_2O)_2POCH_2CO-$ ,

$(\text{CH}_3\text{CH}_2\text{O})_2\text{POCH}_2\text{CH}_2\text{CO}-$ ,  $(\text{CH}_3\text{O})_2\text{POCH}_2\text{CH}_2\text{CO}-$ ,  $(\text{CH}_3\text{CH}_2\text{CH}_2\text{CH}_2\text{O})_2\text{POCH}_2\text{CH}_2\text{CO}-$ ,  
 $(\text{CH}_3\text{CH}_2\text{O})_2\text{PO}(\text{CH}_2)_4\text{CO}-$ ,  $(\text{CH}_3\text{CH}_2\text{O})_2\text{PO}(\text{CH}_2)_8\text{CO}-$  or  $(\text{CH}_3\text{CH}_2\text{O})_2\text{PO}(\text{CH}_2)_{17}\text{CO}-$ .

Alkanoyloxy having up to 25 carbon atoms is a branched or unbranched radical, for example formyloxy, acetoxy, propionyloxy, butanoyloxy, pentanoyloxy, hexanoyloxy, heptanoyloxy, octanoyloxy, nonanoyloxy, decanoyloxy, undecanoyloxy, dodecanoyloxy, tridecanoyloxy, tetradecanoyloxy, pentadecanoyloxy, hexadecanoyloxy, heptadecanoyloxy, octadecanoyloxy, eicosanoyloxy or docosanoyloxy. Preference is given to alkanoyloxy having 2 to 18, especially 2 to 12, for example 2 to 6 carbon atoms. Particular preference is given to acetoxy.

Alkenoyl having 3 to 25 carbon atoms is a branched or unbranched radical, for example propenoyl, 2-butenoyl, 3-butenoyl, isobutenoyl, n-2,4-pentadienoyl, 3-methyl-2-butenoyl, n-2-octenoyl, n-2-dodecenoyl, iso-dodecenoyl, oleoyl, n-2-octadecenoyl or n-4-octadecenoyl. Preference is given to alkenoyl having 3 to 18, especially 3 to 12, for example 3 to 6, in particular 3 to 4 carbon atoms.

$\text{C}_3\text{-C}_{25}$ alkenoyl interrupted by oxygen, sulfur or  $\text{>N-R}_{14}$  is, for example,

$\text{CH}_3\text{OCH}_2\text{CH}_2\text{CH}=\text{CHCO}-$  or  $\text{CH}_3\text{OCH}_2\text{CH}_2\text{OCH}=\text{CHCO}-$ .

Alkenoyloxy having 3 to 25 carbon atoms is a branched or unbranched radical, for example propenoyloxy, 2-butenoyloxy, 3-butenoyloxy, isobutenoyloxy, n-2,4-pentadienoyloxy, 3-methyl-2-butenoyloxy, n-2-octenoyloxy, n-2-dodecenoyloxy, iso-dodecenoyloxy, oleoyloxy, n-2-octadecenoyloxy or n-4-octadecenoyloxy. Preference is given to alkenoyloxy having 3 to 18, especially 3 to 12, for example 3 to 6, in particular 3 to 4 carbon atoms.

$\text{C}_3\text{-C}_{25}$ alkenoyloxy interrupted by oxygen, sulfur or  $\text{>N-R}_{14}$  is, for example,

$\text{CH}_3\text{OCH}_2\text{CH}_2\text{CH}=\text{CHCOO}-$  or  $\text{CH}_3\text{OCH}_2\text{CH}_2\text{OCH}=\text{CHCOO}-$ .

$C_3-C_{25}$ alkanoyl interrupted by oxygen, sulfur or  $\text{>N}-R_{14}$  is, for example,  $\text{CH}_3\text{-O-CH}_2\text{CO-}$ ,  $\text{CH}_3\text{-S-CH}_2\text{CO-}$ ,  $\text{CH}_3\text{-NH-CH}_2\text{CO-}$ ,  $\text{CH}_3\text{-N(CH}_3\text{)-CH}_2\text{CO-}$ ,  $\text{CH}_3\text{-O-CH}_2\text{CH}_2\text{-O-CH}_2\text{CO-}$ ,  $\text{CH}_3\text{-(O-CH}_2\text{CH}_2\text{)}_2\text{O-CH}_2\text{CO-}$ ,  $\text{CH}_3\text{-(O-CH}_2\text{CH}_2\text{)}_3\text{O-CH}_2\text{CO-}$  or  $\text{CH}_3\text{-(O-CH}_2\text{CH}_2\text{)}_4\text{O-CH}_2\text{CO-}$ .

$C_3-C_{25}$ alkanoyloxy interrupted by oxygen, sulfur or  $\text{>N}-R_{14}$  is, for example,

$\text{CH}_3\text{-O-CH}_2\text{COO-}$ ,  $\text{CH}_3\text{-S-CH}_2\text{COO-}$ ,  $\text{CH}_3\text{-NH-CH}_2\text{COO-}$ ,  $\text{CH}_3\text{-N(CH}_3\text{)-CH}_2\text{COO-}$ ,  $\text{CH}_3\text{-O-CH}_2\text{CH}_2\text{-O-CH}_2\text{COO-}$ ,  $\text{CH}_3\text{-(O-CH}_2\text{CH}_2\text{)}_2\text{O-CH}_2\text{COO-}$ ,  $\text{CH}_3\text{-(O-CH}_2\text{CH}_2\text{)}_3\text{O-CH}_2\text{COO-}$  or  $\text{CH}_3\text{-(O-CH}_2\text{CH}_2\text{)}_4\text{O-CH}_2\text{COO-}$ .

$C_6-C_9$ cycloalkylcarbonyl is, for example, cyclohexylcarbonyl, cycloheptylcarbonyl or cyclooctylcarbonyl. Cyclohexylcarbonyl is preferred.

$C_6-C_9$ cycloalkylcarbonyloxy is, for example, cyclohexylcarbonyloxy, cycloheptylcarbonyloxy or cyclooctylcarbonyloxy. Cyclohexylcarbonyloxy is preferred.

$C_1-C_{12}$ alkyl-substituted benzoyl, which preferably carries 1 to 3, especially 1 or 2 alkyl groups, is, for example, o-, m- or p-methylbenzoyl, 2,3-dimethylbenzoyl, 2,4-dimethylbenzoyl, 2,5-dimethylbenzoyl, 2,6-dimethylbenzoyl, 3,4-dimethylbenzoyl, 3,5-dimethylbenzoyl, 2-methyl-6-ethylbenzoyl, 4-tert-butylbenzoyl, 2-ethylbenzoyl, 2,4,6-trimethylbenzoyl, 2,6-dimethyl-4-tert-butylbenzoyl or 3,5-di-tert-butylbenzoyl. Preferred substituents are  $C_1-C_8$ alkyl, especially  $C_1-C_4$ alkyl.

$C_1-C_{12}$ alkyl-substituted benzoyloxy, which preferably carries 1 to 3, especially 1 or 2 alkyl groups, is, for example, o-, m- or p-methylbenzoyloxy, 2,3-dimethylbenzoyloxy, 2,4-dimethylbenzoyloxy, 2,5-dimethylbenzoyloxy, 2,6-dimethylbenzoyloxy, 3,4-dimethylbenzoyloxy, 3,5-dimethylbenzoyloxy, 2-methyl-6-ethylbenzoyloxy, 4-tert-butylbenzoyloxy, 2-ethylbenzoyloxy, 2,4,6-trimethylbenzoyloxy, 2,6-dimethyl-4-tert-butylbenzoyloxy or 3,5-di-tert-butylbenzoyloxy. Preferred substituents are  $C_1-C_8$ alkyl, especially  $C_1-C_4$ alkyl.

Alkyl having up to 25 carbon atoms is a branched or unbranched radical, for example methyl, ethyl, propyl, isopropyl, n-butyl, sec-butyl, isobutyl, tert-butyl, 2-ethylbutyl, n-pentyl,

isopentyl, 1-methylpentyl, 1,3-dimethylbutyl, n-hexyl, 1-methylhexyl, n-heptyl, isoheptyl, 1,1,3,3-tetramethylbutyl, 1-methylheptyl, 3-methylheptyl, n-octyl, 2-ethylhexyl, 1,1,3-trimethylhexyl, 1,1,3,3-tetramethylpentyl, nonyl, decyl, undecyl, 1-methylundecyl, dodecyl, 1,1,3,3,5,5-hexamethylhexyl, tridecyl, tetradecyl, pentadecyl, hexadecyl, heptadecyl, octadecyl, eicosyl or docosyl. One of the preferred meanings of  $R_2$  and  $R_4$  is, for example,  $C_1$ - $C_{18}$ alkyl. A particularly preferred meaning of  $R_4$  is  $C_1$ - $C_4$ alkyl.

Alkenyl having 3 to 25 carbon atoms is a branched or unbranched radical, for example propenyl, 2-butenyl, 3-butenyl, isobutenyl, n-2,4-pentadienyl, 3-methyl-2-butenyl, n-2-octenyl, n-2-dodecenyl, iso-dodecenyl, oleyl, n-2-octadecenyl or n-4-octadecenyl. Preference is given to alkenyl having 3 to 18, especially 3 to 12, for example 3 to 6, in particular 3 to 4 carbon atoms.

Alkenyloxy having 3 to 25 carbon atoms is a branched or unbranched radical, for example propenyloxy, 2-butenyloxy, 3-butenyloxy, isobutenyloxy, n-2,4-pentadienyloxy, 3-methyl-2-butenyloxy, n-2-octenyloxy, n-2-dodecenyloxy, iso-dodecenyloxy, oleyloxy, n-2-octadecenyloxy or n-4-octadecenyloxy. Preference is given to alkenyloxy having 3 to 18, especially 3 to 12, for example 3 to 6, in particular 3 to 4 carbon atoms.

Alkynyl having 3 to 25 carbon atoms is a branched or unbranched radical, for example propynyl (  $-\text{CH}_2-\text{C}\equiv\text{CH}$  ), 2-butylnyl, 3-butylnyl, n-2-octynyl, or n-2-dodecynyl. Preference is given to alkynyl having 3 to 18, especially 3 to 12, for example 3 to 6, in particular 3 to 4 carbon atoms.

Alkynyloxy having 3 to 25 carbon atoms is a branched or unbranched radical, for example propynyloxy (  $-\text{OCH}_2-\text{C}\equiv\text{CH}$  ), 2-butyntyloxy, 3-butyntyloxy, n-2-octynyloxy, or n-2-dodecynyloxy. Preference is given to alkynyloxy having 3 to 18, especially 3 to 12, for example 3 to 6, in particular 3 to 4 carbon atoms.

C<sub>2</sub>-C<sub>25</sub>alkyl interrupted by oxygen, sulfur or  $\text{>N-R}_{14}$  is, for example, CH<sub>3</sub>-O-CH<sub>2</sub>-

CH<sub>3</sub>-S-CH<sub>2</sub>-, CH<sub>3</sub>-NH-CH<sub>2</sub>-, CH<sub>3</sub>-N(CH<sub>3</sub>)-CH<sub>2</sub>-, CH<sub>3</sub>-O-CH<sub>2</sub>CH<sub>2</sub>-O-CH<sub>2</sub>-,  
CH<sub>3</sub>-(O-CH<sub>2</sub>CH<sub>2</sub>)<sub>2</sub>O-CH<sub>2</sub>-, CH<sub>3</sub>-(O-CH<sub>2</sub>CH<sub>2</sub>)<sub>3</sub>O-CH<sub>2</sub>- or CH<sub>3</sub>-(O-CH<sub>2</sub>CH<sub>2</sub>)<sub>4</sub>O-CH<sub>2</sub>-.

C<sub>7</sub>-C<sub>9</sub>phenylalkyl is, for example, benzyl, α-methylbenzyl, α,α-dimethylbenzyl or 2-phenylethyl. Benzyl and α,α-dimethylbenzyl are preferred.

C<sub>7</sub>-C<sub>9</sub>phenylalkyl which is unsubstituted or substituted on the phenyl radical from 1 to 3 times by C<sub>1</sub>-C<sub>4</sub>alkyl is, for example, benzyl, α-methylbenzyl, α,α-dimethylbenzyl, 2-phenylethyl, 2-methylbenzyl, 3-methylbenzyl, 4-methylbenzyl, 2,4-dimethylbenzyl, 2,6-dimethylbenzyl or 4-tert-butylbenzyl. Benzyl is preferred.

C<sub>7</sub>-C<sub>25</sub>phenylalkyl which is unsubstituted or substituted on the phenyl radical from 1 to 3 times by C<sub>1</sub>-C<sub>4</sub>alkyl and is interrupted by oxygen, sulfur or  $\text{>N-R}_{14}$  is a branched or un-

branched radical, for example phenoxymethyl, 2-methylphenoxymethyl, 3-methylphenoxy-methyl, 4-methylphenoxymethyl, 2,4-dimethylphenoxymethyl, 2,3-dimethylphenoxymethyl, phenylthiomethyl, N-methyl-N-phenylmethyl, N-ethyl-N-phenylmethyl, 4-tert-butylphenoxy-methyl, 4-tert-butylphenoxyethoxymethyl, 2,4-di-tert-butylphenoxyethoxymethyl, 2,4-di-tert-butyl-phenoxyethoxymethyl, phenoxyethoxyethoxyethoxymethyl, benzyloxymethyl, benzyloxy-ethoxymethyl, N-benzyl-N-ethylmethyl or N-benzyl-N-isopropylmethyl.

C<sub>7</sub>-C<sub>9</sub>phenylalkoxy is, for example, benzyloxy, α-methylbenzyloxy, α,α-dimethylbenzyloxy or 2-phenylethoxy. Benzyloxy is preferred.

C<sub>1</sub>-C<sub>4</sub>alkyl-substituted phenyl, which preferably contains 1 to 3, especially 1 or 2 alkyl groups, is, for example, o-, m- or p-methylphenyl, 2,3-dimethylphenyl, 2,4-dimethylphenyl, 2,5-dimethylphenyl, 2,6-dimethylphenyl, 3,4-dimethylphenyl, 3,5-dimethylphenyl, 2-methyl-6-ethylphenyl, 4-tert-butylphenyl, 2-ethylphenyl or 2,6-diethylphenyl.

C<sub>1</sub>-C<sub>4</sub>alkyl-substituted phenoxy, which preferably contains 1 to 3, especially 1 or 2 alkyl groups, is, for example, o-, m- or p-methylphenoxy, 2,3-dimethylphenoxy, 2,4-dimethylphe-

noxy, 2,5-dimethylphenoxy, 2,6-dimethylphenoxy, 3,4-dimethylphenoxy, 3,5-dimethylphenoxy, 2-methyl-6-ethylphenoxy, 4-tert-butylphenoxy, 2-ethylphenoxy or 2,6-diethylphenoxy.

Unsubstituted or C<sub>1</sub>-C<sub>4</sub>alkyl-substituted C<sub>5</sub>-C<sub>8</sub>cycloalkyl is, for example, cyclopentyl, methylcyclopentyl, dimethylcyclopentyl, cyclohexyl, methylcyclohexyl, dimethylcyclohexyl, trimethylcyclohexyl, tert-butylcyclohexyl, cycloheptyl or cyclooctyl. Preference is given to cyclohexyl and tert-butylcyclohexyl.

Unsubstituted or C<sub>1</sub>-C<sub>4</sub>alkyl-substituted C<sub>5</sub>-C<sub>8</sub>cycloalkoxy is, for example, cyclopentoxy, methylcyclopentoxy, dimethylcyclopentoxy, cyclohexoxy, methylcyclohexoxy, dimethylcyclohexoxy, trimethylcyclohexoxy, tert-butylcyclohexoxy, cycloheptoxy or cyclooctoxy. Preference is given to cyclohexoxy and tert-butylcyclohexoxy.

Alkoxy having up to 25 carbon atoms is a branched or unbranched radical, for example methoxy, ethoxy, propoxy, isopropoxy, n-butoxy, isobutoxy, pentoxy, isopentoxy, hexoxy, heptoxy, octoxy, decyloxy, tetradecyloxy, hexadecyloxy or octadecyloxy. Preference is given to alkoxy having 1 to 12, especially 1 to 8, for example 1 to 6 carbon atoms.

C<sub>2</sub>-C<sub>25</sub>alkoxy interrupted by oxygen, sulfur or  $\text{>N-R}_{14}$  is, for example, CH<sub>3</sub>-O-CH<sub>2</sub>CH<sub>2</sub>O-

CH<sub>3</sub>-S-CH<sub>2</sub>CH<sub>2</sub>O-, CH<sub>3</sub>-NH-CH<sub>2</sub>CH<sub>2</sub>O-, CH<sub>3</sub>-N(CH<sub>3</sub>)-CH<sub>2</sub>CH<sub>2</sub>O-,  
CH<sub>3</sub>-O-CH<sub>2</sub>CH<sub>2</sub>-O-CH<sub>2</sub>CH<sub>2</sub>O-, CH<sub>3</sub>-(O-CH<sub>2</sub>CH<sub>2</sub>)<sub>2</sub>O-CH<sub>2</sub>CH<sub>2</sub>O-,  
CH<sub>3</sub>-(O-CH<sub>2</sub>CH<sub>2</sub>)<sub>3</sub>O-CH<sub>2</sub>CH<sub>2</sub>O- or CH<sub>3</sub>-(O-CH<sub>2</sub>CH<sub>2</sub>)<sub>4</sub>O-CH<sub>2</sub>CH<sub>2</sub>O-.

Alkylthio having up to 25 carbon atoms is a branched or unbranched radical, for example methylthio, ethylthio, propylthio, isopropylthio, n-butylthio, isobutylthio, pentylthio, isopentylthio, hexylthio, heptylthio, octylthio, decylthio, tetradecylthio, hexadecylthio or octadecylthio. Preference is given to alkylthio having 1 to 12, especially 1 to 8, for example 1 to 6 carbon atoms.

Alkylamino having up to 4 carbon atoms is a branched or unbranched radical, for example methylamino, ethylamino, propylamino, isopropylamino, n-butylamino, isobutylamino or tert-butylamino.

Di(C<sub>1</sub>-C<sub>4</sub>alkyl)amino also means that the two radicals independently of one another are branched or unbranched, , for example dimethylamino, methylethylamino, diethylamino, methyl-n-propylamino, methylisopropylamino, methyl-n-butylamino, methylisobutylamino, ethylisopropylamino, ethyl-n-butylamino, ethylisobutylamino, ethyl-tert-butylamino, diethylamino, diisopropylamino, isopropyl-n-butylamino, isopropylisobutylamino, di-n-butylamino or diisobutylamino.

Alkanoylamino having up to 25 carbon atoms is a branched or unbranched radical , for example formylamino, acetylamino, propionylamino, butanoylamino, pentanoylamino, hexanoylamino, heptanoylamino, octanoylamino, nonanoylamino, decanoylamino, undecanoylamino, dodecanoylamino, tridecanoylamino, tetradecanoylamino, pentadecanoylamino, hexadecanoylamino, heptadecanoylamino, octadecanoylamino, eicosanoylamino or docosanoylamino. Preference is given to alkanoylamino having 2 to 18, especially 2 to 12, for example 2 to 6 carbon atoms.

C<sub>1</sub>-C<sub>18</sub>alkylene is a branched or unbranched radical , for example methylene, ethylene, propylene, trimethylene, tetramethylene, pentamethylene, hexamethylene, heptamethylene, octamethylene, decamethylene, dodecamethylene or octadecamethylene. Preference is given to C<sub>1</sub>-C<sub>12</sub>alkylene, especially C<sub>1</sub>-C<sub>8</sub>alkylene.

A C<sub>1</sub>-C<sub>4</sub>alkyl-substituted C<sub>5</sub>-C<sub>12</sub>cycloalkylene ring, which preferably contains 1 to 3, especially 1 or 2 branched or unbranched alkyl group radicals, is, for example, cyclopentylene, methylcyclopentylene, dimethylcyclopentylene, cyclohexylene, methylcyclohexylene, dimethylcyclohexylene, trimethylcyclohexylene, tert-butylcyclohexylene, cycloheptylene, cyclooctylene or cyclodecylene. Preference is given to cyclohexylene and tert-butylcyclohexylene.

C<sub>2</sub>-C<sub>18</sub>alkylene interrupted by oxygen, sulfur or  $\text{>N-R}_{14}$  is, for example, -CH<sub>2</sub>-O-CH<sub>2</sub>-

-CH<sub>2</sub>-S-CH<sub>2</sub>-, -CH<sub>2</sub>-NH-CH<sub>2</sub>-, -CH<sub>2</sub>-N(CH<sub>3</sub>)-CH<sub>2</sub>-, -CH<sub>2</sub>-O-CH<sub>2</sub>CH<sub>2</sub>-O-CH<sub>2</sub>-,  
-CH<sub>2</sub>-(O-CH<sub>2</sub>CH<sub>2</sub>)<sub>2</sub>-O-CH<sub>2</sub>-, -CH<sub>2</sub>-(O-CH<sub>2</sub>CH<sub>2</sub>)<sub>3</sub>-O-CH<sub>2</sub>-, -CH<sub>2</sub>-(O-CH<sub>2</sub>CH<sub>2</sub>)<sub>4</sub>-O-CH<sub>2</sub>- or  
-CH<sub>2</sub>CH<sub>2</sub>-S-CH<sub>2</sub>CH<sub>2</sub>-.



C<sub>2</sub>-C<sub>18</sub>alkenylene is, for example, vinylene, methylvinylene, octenylethylene or dodecenylethylene. Preference is given to C<sub>2</sub>-C<sub>8</sub>alkenylene.

Alkylidene having 2 to 20 carbon atoms is, for example, ethylidene, propylidene, butylidene, pentylidene, 4-methylpentylidene, heptylidene, nonylidene, tridecylidene, nonadecylidene, 1-methylethylidene, 1-ethylpropylidene or 1-ethylpentylidene. Preference is given to C<sub>2</sub>-C<sub>8</sub>alkylidene.

Phenylalkylidene having 7 to 20 carbon atoms is, for example, benzylidene, 2-phenylethylidene or 1-phenyl-2-hexylidene. Preference is given to C<sub>7</sub>-C<sub>9</sub>-phenylalkylidene.

C<sub>5</sub>-C<sub>8</sub>cycloalkylene is a saturated hydrocarbon group having two free valencies and at least one ring unit and is, for example, cyclopentylene, cyclohexylene, cycloheptylene or cyclooctylene. Preference is given to cyclohexylene.

C<sub>7</sub>-C<sub>8</sub>bicycloalkylene is, for example, bicycloheptylene or bicyclooctylene.

Unsubstituted or C<sub>1</sub>-C<sub>4</sub>alkyl-substituted phenylene or naphthylene is, for example, 1,2-, 1,3-, 1,4-phenylene, 1,2-, 1,3-, 1,4-, 1,6-, 1,7-, 2,6- or 2,7-naphthylene. 1,4-Phenylene is preferred.

A C<sub>1</sub>-C<sub>4</sub>alkyl-substituted C<sub>5</sub>-C<sub>8</sub>cycloalkylidene ring, which preferably contains 1 to 3, especially 1 or 2 branched or unbranched alkyl group radicals, is, for example, cyclopentylidene, methylcyclopentylidene, dimethylcyclopentylidene, cyclohexylidene, methylcyclohexylidene, dimethylcyclohexylidene, trimethylcyclohexylidene, tert-butylcyclohexylidene, cycloheptylidene or cyclooctylidene. Preference is given to cyclohexylidene and tert-butylcyclohexylidene.

A mono-, di- or trivalent metal cation is preferably an alkali metal, alkaline earth metal or aluminium cation, for example, Na<sup>+</sup>, K<sup>+</sup>, Mg<sup>++</sup>, Ca<sup>++</sup> or Al<sup>+++</sup>.

Compositions that are of interest include those comprising as component (b) at least one compound of the formula I in which, if n is 1, R<sub>1</sub> is unsubstituted phenyl or phenyl which is substituted in the para position by C<sub>1</sub>-C<sub>18</sub>alkylthio or di(C<sub>1</sub>-C<sub>4</sub>alkyl)amino; mono- to penta-

substituted alkylphenyl having in total together not more than 18 carbon atoms in the 1 to 5 alkyl substituents; or is unsubstituted or C<sub>1</sub>-C<sub>4</sub>alkyl-, C<sub>1</sub>-C<sub>4</sub>alkoxy-, C<sub>1</sub>-C<sub>4</sub>alkylthio-, hydroxyl- or amino-substituted naphthyl, biphenyl, terphenyl, phenanthryl, anthryl, fluorenyl, carbazoyl, thienyl, pyrrolyl, phenothiazinyl or 5,6,7,8-tetrahydronaphthyl.

Preference is given to compositions comprising as component (b) at least one compound of the formula I in which, if n is 2,

R<sub>1</sub> is -R<sub>12</sub>-X-R<sub>13</sub>-,

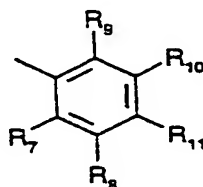
R<sub>12</sub> and R<sub>13</sub> are phenylene,

X is oxygen or -NR<sub>31</sub>-, and

R<sub>31</sub> is C<sub>1</sub>-C<sub>4</sub>alkyl.

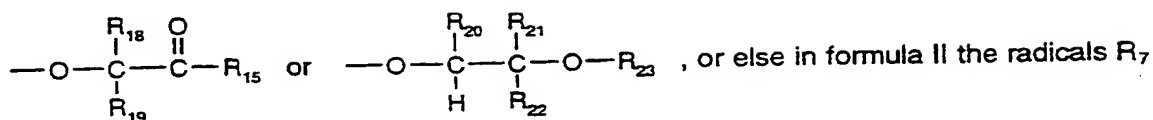
Preference is also given to compositions comprising as component (b) at least one compound of the formula I in which, if n is 1,

R<sub>1</sub> is unsubstituted or C<sub>1</sub>-C<sub>4</sub>alkyl-, C<sub>1</sub>-C<sub>4</sub>alkoxy-, C<sub>1</sub>-C<sub>4</sub>alkylthio-, hydroxyl-, halo-, amino-, C<sub>1</sub>-C<sub>4</sub>alkylamino- or di(C<sub>1</sub>-C<sub>4</sub>alkyl)amino-substituted naphthyl, phenanthryl, thienyl, dibenzofuryl, carbazoyl, fluorenyl or a radical of the formula II



(II)

R<sub>7</sub>, R<sub>8</sub>, R<sub>9</sub>, R<sub>10</sub> and R<sub>11</sub> independently of one another are hydrogen, chlorine, bromine, hydroxyl, C<sub>1</sub>-C<sub>18</sub>alkyl, C<sub>2</sub>-C<sub>18</sub>alkyl interrupted by oxygen or sulfur, C<sub>1</sub>-C<sub>18</sub>alkoxy, C<sub>2</sub>-C<sub>18</sub>alkoxy interrupted by oxygen or sulfur, C<sub>1</sub>-C<sub>18</sub>alkylthio, C<sub>3</sub>-C<sub>12</sub>alkenylloxy, C<sub>3</sub>-C<sub>12</sub>alkynylloxy, C<sub>7</sub>-C<sub>9</sub>phenylalkyl, C<sub>7</sub>-C<sub>9</sub>phenylalkoxy, unsubstituted or C<sub>1</sub>-C<sub>4</sub>alkyl-substituted phenyl; phenoxy, cyclohexyl, C<sub>5</sub>-C<sub>8</sub>cycloalkoxy, C<sub>1</sub>-C<sub>4</sub>alkylamino, di(C<sub>1</sub>-C<sub>4</sub>alkyl)amino, C<sub>1</sub>-C<sub>12</sub>alkanoyl, C<sub>3</sub>-C<sub>12</sub>alkanoyl interrupted by oxygen or sulfur, C<sub>1</sub>-C<sub>12</sub>alkanoyloxy, C<sub>3</sub>-C<sub>12</sub>alkanoyloxy interrupted by oxygen or sulfur, C<sub>1</sub>-C<sub>12</sub>alkanoylamino, C<sub>3</sub>-C<sub>12</sub>alkenoyl, C<sub>3</sub>-C<sub>12</sub>alkenoyloxy, cyclohexylcarbonyl, cyclohexylcarbonyloxy, benzoyl or C<sub>1</sub>-C<sub>4</sub>alkyl-substituted benzoyl; benzoyloxy or C<sub>1</sub>-C<sub>4</sub>alkyl-substituted benzoyloxy;



and R<sub>8</sub> or the radicals R<sub>8</sub> and R<sub>11</sub>, together with the carbon atoms to which they are attached, form a benzo ring,



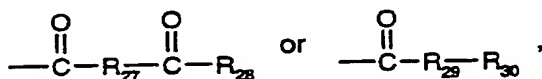
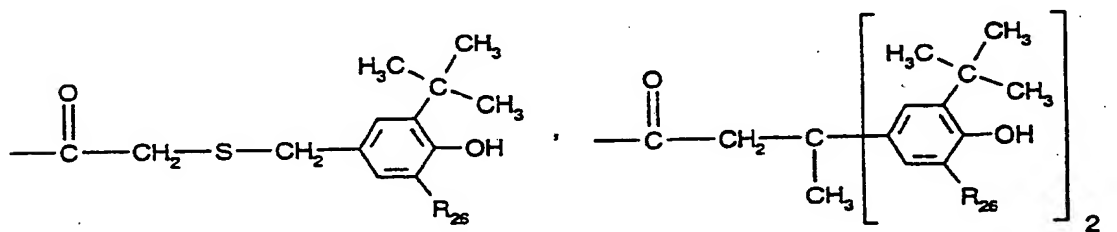
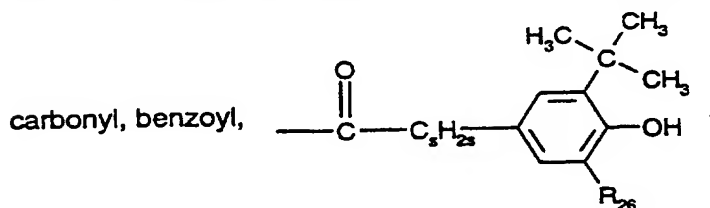
R<sub>18</sub> and R<sub>19</sub> independently of one another are hydrogen or C<sub>1</sub>-C<sub>4</sub>alkyl,

R<sub>20</sub> is hydrogen,

R<sub>21</sub> is hydrogen, phenyl, C<sub>1</sub>-C<sub>18</sub>alkyl, C<sub>2</sub>-C<sub>18</sub>alkyl interrupted by oxygen or sulfur; C<sub>7</sub>-C<sub>9</sub>phenylalkyl, C<sub>7</sub>-C<sub>18</sub>-phenylalkyl which is unsubstituted or substituted on the phenyl radical from 1 to 3 times by C<sub>1</sub>-C<sub>4</sub>alkyl and is interrupted by oxygen or sulfur, or else the radicals R<sub>20</sub> and R<sub>21</sub>, together with the carbon atoms to which they are attached, form a cyclohexylene ring which is unsubstituted or substituted from 1 to 3 times by C<sub>1</sub>-C<sub>4</sub>alkyl,

R<sub>22</sub> is hydrogen or C<sub>1</sub>-C<sub>4</sub>alkyl,

R<sub>23</sub> is hydrogen, C<sub>1</sub>-C<sub>18</sub>alkanoyl, C<sub>3</sub>-C<sub>18</sub>alkenoyl, C<sub>3</sub>-C<sub>12</sub>alkanoyl interrupted by oxygen or sulfur; C<sub>2</sub>-C<sub>12</sub>alkanoyl substituted by a di(C<sub>1</sub>-C<sub>6</sub>-alkyl)phosphonate group; C<sub>6</sub>-C<sub>9</sub>cycloalkyl-



R<sub>24</sub> and R<sub>25</sub> independently of one another are hydrogen or C<sub>1</sub>-C<sub>12</sub>alkyl,

R<sub>26</sub> is hydrogen or C<sub>1</sub>-C<sub>4</sub>alkyl,

R<sub>27</sub> is C<sub>1</sub>-C<sub>12</sub>alkylene, C<sub>2</sub>-C<sub>8</sub>alkenylene, C<sub>2</sub>-C<sub>8</sub>alkylidene, C<sub>7</sub>-C<sub>12</sub>phenylalkylidene, C<sub>5</sub>-C<sub>8</sub>cycloalkylene or phenylene,

R<sub>28</sub> is hydroxyl, C<sub>1</sub>-C<sub>12</sub>alkoxy or  $\text{---N} \begin{matrix} \text{R}_{24} \\ \text{R}_{25} \end{matrix}$ ,

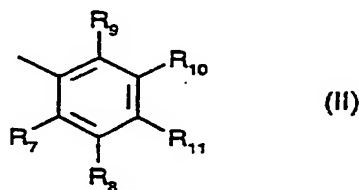
R<sub>29</sub> is oxygen or -NH-,

R<sub>30</sub> is C<sub>1</sub>-C<sub>18</sub>alkyl or phenyl, and

s is 1 or 2.

Preference is likewise given to compositions comprising as component (b) at least one compound of the formula I in which, if n is 1,

R<sub>1</sub> is phenanthryl, thienyl, dibenzofuryl, unsubstituted or C<sub>1</sub>-C<sub>4</sub>alkyl-substituted carbazoyl; or is fluorenyl; or R<sub>1</sub> is a radical of the formula II



R<sub>7</sub>, R<sub>8</sub>, R<sub>9</sub>, R<sub>10</sub> and R<sub>11</sub> independently of one another are hydrogen, chlorine, hydroxyl, C<sub>1</sub>-C<sub>18</sub>alkyl, C<sub>1</sub>-C<sub>18</sub>alkoxy, C<sub>1</sub>-C<sub>18</sub>alkylthio, C<sub>3</sub>-C<sub>4</sub>alkenyloxy, C<sub>3</sub>-C<sub>4</sub>alkynyloxy, phenyl, benzoyl,

benzoyloxy or  $\text{---O---} \begin{matrix} \text{R}_{20} & \text{R}_{21} \\ | & | \\ \text{C} & \text{---C---} \\ | & | \\ \text{H} & \text{R}_{22} \end{matrix} \text{---O---R}_{23}$ .

R<sub>20</sub> is hydrogen,

R<sub>21</sub> is hydrogen, phenyl or C<sub>1</sub>-C<sub>18</sub>alkyl, or else the radicals R<sub>20</sub> and R<sub>21</sub>, together with the carbon atoms to which they are attached, form a cyclohexylene ring which is unsubstituted or substituted from 1 to 3 times by C<sub>1</sub>-C<sub>4</sub>alkyl,

R<sub>22</sub> is hydrogen or C<sub>1</sub>-C<sub>4</sub>alkyl, and

R<sub>23</sub> is hydrogen, C<sub>1</sub>-C<sub>12</sub>alkanoyl or benzoyl.

Particular preference is given to compositions comprising as component (b) at least one compound of the formula I in which, if n is 1,

R<sub>7</sub>, R<sub>8</sub>, R<sub>9</sub>, R<sub>10</sub> and R<sub>11</sub> independently of one another are hydrogen, C<sub>1</sub>-C<sub>12</sub>alkyl, C<sub>1</sub>-C<sub>4</sub>alkylthio or phenyl.

Of particular interest are compositions comprising as component (b) at least one compound of the formula I in which

R<sub>2</sub>, R<sub>3</sub>, R<sub>4</sub> and R<sub>5</sub> independently of one another are hydrogen, chlorine, C<sub>1</sub>-C<sub>18</sub>alkyl, benzyl, phenyl, C<sub>5</sub>-C<sub>8</sub>cycloalkyl, C<sub>1</sub>-C<sub>18</sub>alkoxy, C<sub>1</sub>-C<sub>18</sub>alkylthio, C<sub>1</sub>-C<sub>18</sub>alkanoyloxy, C<sub>1</sub>-C<sub>18</sub>alkanoyl-amino, C<sub>3</sub>-C<sub>18</sub>alkenoyloxy or benzoyloxy; or else the radicals R<sub>2</sub> and R<sub>3</sub> or the radicals R<sub>3</sub> and R<sub>4</sub> or the radicals R<sub>4</sub> and R<sub>5</sub>, together with the carbon atoms to which they are attached, form a benzo ring, R<sub>4</sub> is additionally -(CH<sub>2</sub>)<sub>p</sub>-COR<sub>15</sub> or -(CH<sub>2</sub>)<sub>q</sub>OH, or, if R<sub>3</sub>, R<sub>5</sub> and R<sub>6</sub> are hydrogen, R<sub>4</sub> is additionally a radical of the formula III,

R<sub>15</sub> is hydroxyl, C<sub>1</sub>-C<sub>12</sub>alkoxy or  $\text{---N} \begin{matrix} \nearrow \text{R}_{24} \\ \searrow \text{R}_{25} \end{matrix}$ ,

R<sub>16</sub> and R<sub>17</sub> are methyl groups or, together with the C atom to which they are attached, form a C<sub>5</sub>-C<sub>8</sub>cycloalkylidene ring which is unsubstituted or substituted from 1 to 3 times by C<sub>1</sub>-C<sub>4</sub>alkyl,

R<sub>24</sub> and R<sub>25</sub> independently of one another are hydrogen or C<sub>1</sub>-C<sub>12</sub>alkyl,

p is 1 or 2, and

q is 2, 3, 4, 5 or 6.

Also of particular interest are compositions comprising as component (b) at least one compound of the formula I in which at least two of the radicals R<sub>2</sub>, R<sub>3</sub>, R<sub>4</sub> and R<sub>5</sub> are hydrogen.

Of special interest are compositions comprising as component (b) at least one compound of the formula I in which R<sub>3</sub> and R<sub>5</sub> are hydrogen.

Of very special interest are compositions comprising as component (b) at least one compound of the formula I in which

R<sub>2</sub> is C<sub>1</sub>-C<sub>4</sub>alkyl,

R<sub>3</sub> is hydrogen,

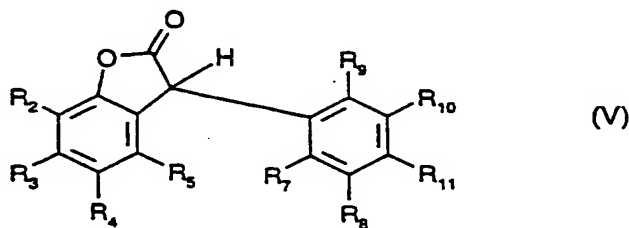
R<sub>4</sub> is C<sub>1</sub>-C<sub>4</sub>alkyl or, if R<sub>6</sub> is hydrogen, R<sub>4</sub> is additionally a radical of the formula III,

R<sub>5</sub> is hydrogen, and

R<sub>16</sub> and R<sub>17</sub>, together with the C atom to which they are attached, form a cyclohexylidene.

The following compounds are examples of the benzofuran-2-one type which are particularly suitable as component (b) in the novel composition: 3-[4-(2-acetoxyethoxy)phenyl]-5,7-di-tert-butyl-benzofuran-2-one; 5,7-di-tert-butyl-3-[4-(2-stearoyloxyethoxy)phenyl]benzofuran-2-one; 3,3'-bis[5,7-di-tert-butyl-3-(4-[2-hydroxyethoxy]phenyl)benzofuran-2-one]; 5,7-di-tert-butyl-3-(4-ethoxyphenyl)benzofuran-2-one; 3-(4-acetoxy-3,5-dimethylphenyl)-5,7-di-tert-butylbenzofuran-2-one; 3-(3,5-dimethyl-4-pivaloyloxy-phenyl)-5,7-di-tert-butyl-benzofuran-2-one; 5,7-di-tert-butyl-3-phenylbenzofuran-2-one; 5,7-di-tert-butyl-3-(3,4-dimethylphenyl)-benzofuran-2-one; 5,7-di-tert-butyl-3-(2,3-dimethylphenyl)benzofuran-2-one.

Also of special interest are compositions comprising as component (b) at least one compound of the formula V



in which

R<sub>2</sub> is hydrogen or C<sub>1</sub>-C<sub>6</sub>alkyl,

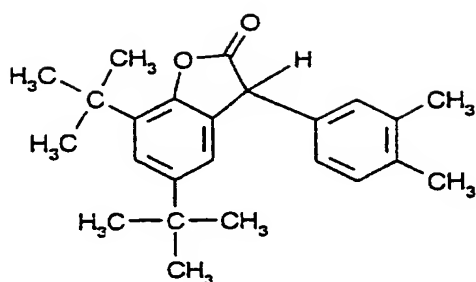
R<sub>3</sub> is hydrogen,

R<sub>4</sub> is hydrogen or C<sub>1</sub>-C<sub>6</sub>alkyl,

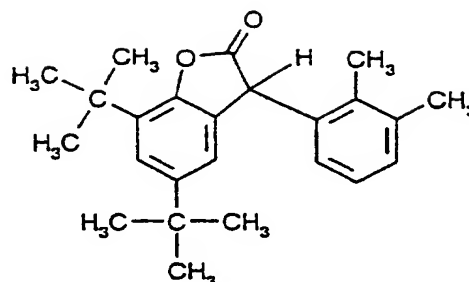
R<sub>5</sub> is hydrogen,

R<sub>7</sub>, R<sub>8</sub>, R<sub>9</sub>, R<sub>10</sub> and R<sub>11</sub> independently of one another are hydrogen, C<sub>1</sub>-C<sub>4</sub>alkyl or C<sub>1</sub>-C<sub>4</sub>alkoxy, with the proviso that at least two of the radicals R<sub>7</sub>, R<sub>8</sub>, R<sub>9</sub>, R<sub>10</sub> or R<sub>11</sub> are hydrogen.

Very particular preference is given to compositions comprising as component (b) at least one compound of the formula Va or Vb



(Va)

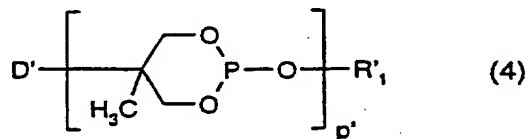
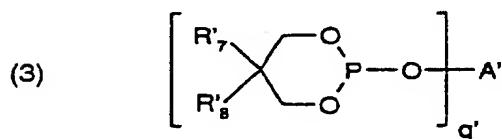
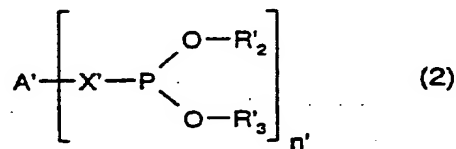
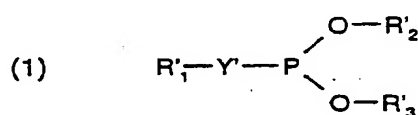


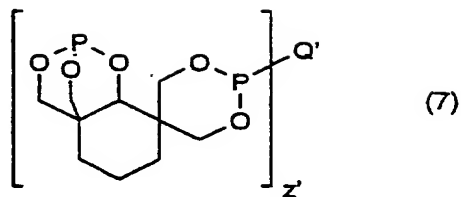
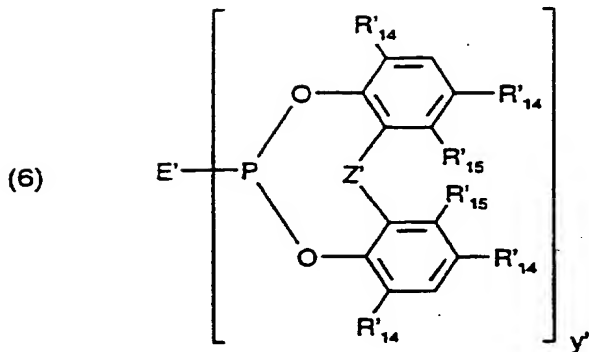
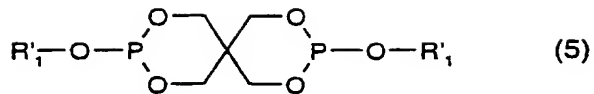
(Vb)

or a mixture of the two compounds of the formula Va and Vb.

The compounds of the benzofuran-2-one type as component (b) in the novel composition are known in the literature and their preparation is described, for example, in the following US patents: US 4,325,863; US 4,388,244; US 5,175,312; US 5,252,643; US 5,216,052; US 5,369,159; US 5,488,117; US 5,356,966; US 5,367,008; US 5,428,162; US 5,428,177 or US 5,516,920.

Of particular interest are compositions comprising as component (c) at least one compound from the group of the organic phosphites or phosphonites of the formulae 1 to 7

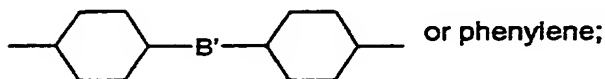
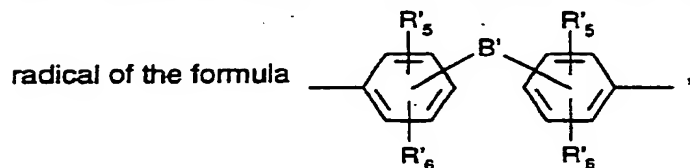




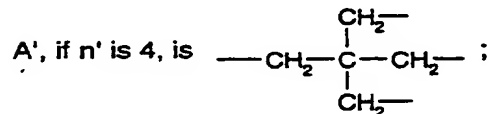
in which the indices are integral and

$n'$  is 2, 3 or 4;  $p'$  is 1 or 2;  $q'$  is 2 or 3;  $r'$  is 4 to 12;  $y'$  is 1, 2 or 3; and  $z'$  is 1 to 6;

$A'$ , if  $n'$  is 2, is  $C_2-C_{18}$ alkylene;  $C_2-C_{12}$ alkylene interrupted by oxygen, sulfur or  $-NR'_4-$ ; a



$A'$ , if  $n'$  is 3, is a radical of the formula  $-C_rH_{2r-1}-$ ;



$A^*$  has the meaning of  $A'$  if  $n'$  is 2;

$B'$  is a direct bond,  $-CH_2-$ ,  $-CHR'_4-$ ,  $-CR'_1R'_4-$ , sulfur or  $C_5-C_7$ cycloalkylidene, or is cyclohexylidene substituted by from 1 to 4  $C_1-C_4$ alkyl radicals in position 3, 4 and/or 5;

$D'$ , if  $p'$  is 1, is methyl and, if  $p'$  is 2, is  $-CH_2OCH_2-$ ;

$E'$ , if  $y'$  is 1, is  $C_1-C_{18}$ alkyl,  $-OR'_1$  or halogen;

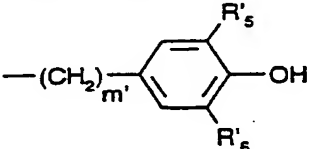
$E'$ , if  $y$  is 2, is  $-O-A^*-O-$ ,



E', if y is 3, is a radical of the formula  $R'_4C(CH_2O-)_3$  or  $N(CH_2CH_2O-)_3$ ;

Q' is the radical of an at least z'-valent alcohol or phenol, this radical being attached via the oxygen atom to the phosphorus atom;

R'<sub>1</sub>, R'<sub>2</sub> and R'<sub>3</sub> independently of one another are unsubstituted or halogen, -COOR'<sub>4</sub>, -CN- or -CONR'<sub>4</sub>R'<sub>4</sub>-substituted C<sub>1</sub>-C<sub>18</sub>alkyl; C<sub>2</sub>-C<sub>18</sub>alkyl interrupted by oxygen, sulfur or -NR'<sub>4</sub>-; C<sub>7</sub>-C<sub>9</sub>phenylalkyl; C<sub>5</sub>-C<sub>12</sub>cycloalkyl, phenyl or naphthyl; naphthyl or phenyl substituted by halogen, 1 to 3 alkyl radicals or alkoxy radicals having in total 1 to 18 carbon atoms or by

C<sub>7</sub>-C<sub>9</sub>phenylalkyl; or are a radical of the formula  in which m' is an

integer from the range 3 to 6;

R'<sub>4</sub> is hydrogen, C<sub>1</sub>-C<sub>18</sub>alkyl, C<sub>5</sub>-C<sub>12</sub>cycloalkyl or C<sub>7</sub>-C<sub>9</sub>phenylalkyl,

R'<sub>5</sub> and R'<sub>6</sub> independently of one another are hydrogen, C<sub>1</sub>-C<sub>8</sub>alkyl or C<sub>5</sub>-C<sub>6</sub>cycloalkyl,

R'<sub>7</sub> and R'<sub>8</sub>, if q' is 2, independently of one another are C<sub>1</sub>-C<sub>4</sub>alkyl or together are a 2,3-dehydropentamethylene radical; and

R'<sub>7</sub> and R'<sub>8</sub>, if q' is 3, are methyl;

R'<sub>14</sub> is hydrogen, C<sub>1</sub>-C<sub>9</sub>alkyl or cyclohexyl,

R'<sub>15</sub> is hydrogen or methyl and, if two or more radicals R'<sub>14</sub> and R'<sub>15</sub> are present, these radicals are identical or different,

X' and Y' are each a direct bond or oxygen,

Z' is a direct bond, methylene, -C(R'<sub>16</sub>)<sub>2</sub>- or sulfur, and

R'<sub>16</sub> is C<sub>1</sub>-C<sub>8</sub>alkyl.

Of particular interest are compositions comprising as component (c) a phosphite or phosphonite of the formula 1, 2, 5 or 6, in which

n' is the number 2 and y' is the number 1, 2 or 3;

A' is C<sub>2</sub>-C<sub>18</sub>alkylene, p-phenylene or p-biphenylene,

E', if y' is 1, is C<sub>1</sub>-C<sub>18</sub>alkyl, -OR'<sub>1</sub> or fluorine;

E', if y' is 2, is p-biphenylene,

E', if y' is 3, is N(CH<sub>2</sub>CH<sub>2</sub>O-)<sub>3</sub>,

R'<sub>1</sub>, R'<sub>2</sub> and R'<sub>3</sub> independently of one another are C<sub>1</sub>-C<sub>18</sub>alkyl, C<sub>7</sub>-C<sub>9</sub>phenylalkyl, cyclohexyl, phenyl, or phenyl substituted by 1 to 3 alkyl radicals having in total 1 to 18 carbon atoms;

R'<sub>14</sub> is hydrogen or C<sub>1</sub>-C<sub>9</sub>alkyl,

R'<sub>15</sub> is hydrogen or methyl;

X' is a direct bond,

Y' is oxygen,

Z' is a direct bond or -CH(R'<sub>16</sub>)-, and

R'<sub>16</sub> is C<sub>1</sub>-C<sub>4</sub>alkyl.

Likewise of interest are compositions comprising as component (c) a phosphite or phosphonite of the formula 1, 2, 5 or 6, in which

n' is the number 2 and y' is the number 1 or 3;

A' is p-biphenylene,

E', if y' is 1, is C<sub>1</sub>-C<sub>18</sub>alkoxy or fluorine,

E', if y' is 3, is N(CH<sub>2</sub>CH<sub>2</sub>O-)<sub>3</sub>,

R'<sub>1</sub>, R'<sub>2</sub> and R'<sub>3</sub> independently of one another are C<sub>1</sub>-C<sub>18</sub>alkyl, or are phenyl substituted by 2 or 3 alkyl radicals having in total 2 to 12 carbon atoms;

R'<sub>14</sub> is methyl or tert-butyl;

R'<sub>15</sub> is hydrogen;

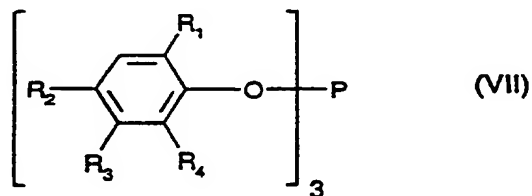
X' is a direct bond;

Y' is oxygen; and

Z' is a direct bond, methylene or -CH(CH<sub>3</sub>)-.

Particular preference is given to compositions comprising as component (c) a phosphite or phosphonite of the formula 1, 2 or 6.

Special preference is given to compositions comprising as component (c) at least one compound of the formula VII



in which

R<sub>1</sub> and R<sub>2</sub> independently of one another are hydrogen, C<sub>1</sub>-C<sub>8</sub>alkyl, cyclohexyl or phenyl, and

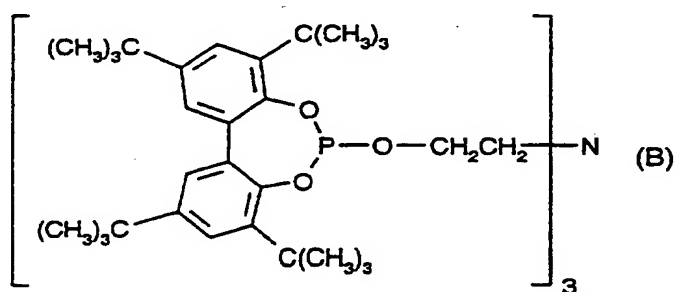
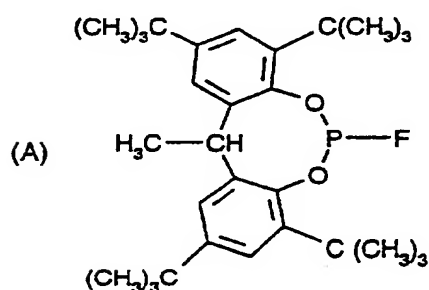
$R_3$  and  $R_4$  independently of one another are hydrogen or  $C_1$ - $C_4$ alkyl.

The following compounds are examples of organic phosphites and phosphonites which are particularly suitable as component (c) in the novel compositions.

Triphenyl phosphite, diphenyl alkyl phosphites, phenyl dialkyl phosphites, tris(nonylphenyl) phosphite, trilauryl phosphite, trioctadecyl phosphite, distearyl pentaerythritol diphosphite, tris(2,4-di-tert-butylphenyl) phosphite (Irgafos® 168, Ciba-Geigy), diisodecyl pentaerythritol diphosphite, bis(2,4-di-tert-butylphenyl) pentaerythritol diphosphite (formula D), bis(2,6-di-tert-butyl-4-methylphenyl) pentaerythritol diphosphite (formula E), bisisodecyloxy-pentaerythritol diphosphite, bis(2,4-di-tert-butyl-6-methylphenyl) pentaerythritol diphosphite, bis(2,4,6-tri-tert-butylphenyl) pentaerythritol diphosphite, tristearyl sorbitol triphosphite, tetrakis(2,4-di-tert-butylphenyl) 4,4'-biphenylenediphosphonite (Irgafos® PEP-Q, Ciba-Geigy, formula H), 6-isoctyloxy-2,4,8,10-tetra-tert-butyl-12H-dibenzo[d,g]-1,3,2-dioxaphosphocin (formula C), 6-fluoro-2,4,8,10-tetra-tert-butyl-12-methyldibenzo[d,g]-1,3,2-dioxaphosphocin (formula A), bis(2,4-di-tert-butyl-6-methylphenyl) methyl phosphite, bis(2,4-di-tert-butyl-6-methylphenyl) ethyl phosphite (formula G).

Particular preference is given to the use of the following phosphites and phosphonites:

tris(2,4-di-tert-butylphenyl) phosphite (Irgafos® 168, Ciba-Geigy), tris(nonylphenyl) phosphite,

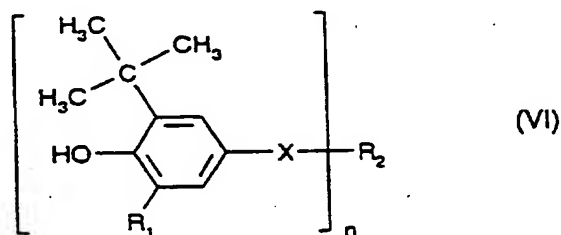




Ciba Spezialitätenchemie AG, formula (G)], or tetrakis(2,4-di-tert-butylphenyl)-4,4'-biphenylenediphosphonite [Irgafos®PEP-Q, Ciba Spezialitätenchemie AG, formula (H)].

The abovementioned organic phosphites and phosphonites are known compounds; many of them are available commercially.

Compositions which are of interest include those comprising as component (d) at least one compound of the formula VI



in which

$\text{R}_1$  is  $\text{C}_1$ - $\text{C}_4$ alkyl,

$n$  is 1, 2, 3 or 4,

$\text{X}$  is methylene,  $-\text{CH}_2-\text{CH}_2-\overset{\text{O}}{\parallel}{\text{C}}-\text{Y}-$  or  $-\text{CH}_2-\overset{\text{O}}{\parallel}{\text{C}}-\text{O}-\text{CH}_2-\text{CH}_2-$ ,  
 $\text{Y}$  is <sup>oxygen</sup>hydrogen or  $-\text{NH}-$ ; and,

if  $n$  is 1,

$\text{X}$  is  $-\text{CH}_2-\text{CH}_2-\overset{\text{O}}{\parallel}{\text{C}}-\text{Y}-$ , where  $\text{Y}$  is attached to  $\text{R}_2$ , and

$\text{R}_2$  is  $\text{C}_1$ - $\text{C}_{25}$ alkyl; and,

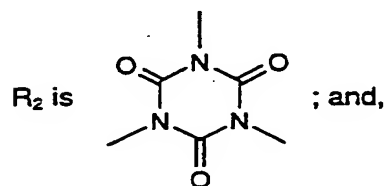
if  $n$  is 2,

X is  $\text{—CH}_2\text{—CH}_2\text{—}\overset{\text{O}}{\parallel}\text{C—Y—}$ , where Y is attached to R<sub>2</sub>, and

R<sub>2</sub> is C<sub>2</sub>–C<sub>12</sub>alkylene, C<sub>4</sub>–C<sub>12</sub>alkylene interrupted by oxygen or sulfur; or, if Y is –NH–, R<sub>2</sub> is additionally a direct bond; and,

if n is 3,

X is methylene or  $\text{—CH}_2\text{—}\overset{\text{O}}{\parallel}\text{C—O—CH}_2\text{—CH}_2\text{—}$ , where the ethylene group is attached to R<sub>2</sub>, and



if n is 4,

X is  $\text{—CH}_2\text{—CH}_2\text{—}\overset{\text{O}}{\parallel}\text{C—Y—}$ , where Y is attached to R<sub>2</sub>, and

R<sub>2</sub> is C<sub>4</sub>–C<sub>10</sub>alkanetetrayl.

Alkyl having up to 25 carbon atoms is a branched or unbranched radical, for example methyl, ethyl, propyl, isopropyl, n-butyl, sec-butyl, isobutyl, tert-butyl, 2-ethylbutyl, n-pentyl, isopentyl, 1-methylpentyl, 1,3-dimethylbutyl, n-hexyl, 1-methylhexyl, n-heptyl, isoheptyl, 1,1,3,3-tetramethylbutyl, 1-methylheptyl, 3-methylheptyl, n-octyl, 2-ethylhexyl, 1,1,3-trimethylhexyl, 1,1,3,3-tetramethylpentyl, nonyl, decyl, undecyl, 1-methylundecyl, dodecyl, 1,1,3,3,5,5-hexamethylhexyl, tridecyl, tetradecyl, pentadecyl, hexadecyl, heptadecyl, octadecyl, eicosyl or docosyl. A preferred definition of R<sub>1</sub> is methyl and tert-butyl. A particularly preferred definition of R<sub>2</sub> is C<sub>1</sub>–C<sub>20</sub>alkyl, especially C<sub>1</sub>–C<sub>18</sub>alkyl, for example C<sub>4</sub>–C<sub>18</sub>alkyl. An especially preferred definition of R<sub>2</sub> is C<sub>8</sub>–C<sub>18</sub>alkyl, especially C<sub>14</sub>–C<sub>18</sub>alkyl, for example C<sub>18</sub>alkyl.

C<sub>2</sub>-C<sub>12</sub>alkylene is a branched or unbranched radical, for example ethylene, propylene, tetramethylene, pentamethylene, hexamethylene, heptamethylene, octamethylene, decamethylene or dodecamethylene. A preferred definition of R<sub>2</sub> is, for example, C<sub>2</sub>-C<sub>10</sub>alkylene, especially C<sub>2</sub>-C<sub>8</sub>alkylene. An especially preferred definition of R<sub>2</sub> is, for example, C<sub>4</sub>-C<sub>8</sub>alkylene, especially C<sub>4</sub>-C<sub>6</sub>alkylene, for example hexamethylene.

C<sub>4</sub>-C<sub>12</sub>alkylene interrupted by oxygen or sulfur can be interrupted one or more times and is, for example, -CH<sub>2</sub>-O-CH<sub>2</sub>CH<sub>2</sub>-O-CH<sub>2</sub>-, -CH<sub>2</sub>-(O-CH<sub>2</sub>CH<sub>2</sub>)<sub>2</sub>O-CH<sub>2</sub>-, -CH<sub>2</sub>-(O-CH<sub>2</sub>CH<sub>2</sub>)<sub>3</sub>O-CH<sub>2</sub>-, -CH<sub>2</sub>-(O-CH<sub>2</sub>CH<sub>2</sub>)<sub>4</sub>O-CH<sub>2</sub>-, -CH<sub>2</sub>CH<sub>2</sub>-O-CH<sub>2</sub>CH<sub>2</sub>-O-CH<sub>2</sub>CH<sub>2</sub>- or -CH<sub>2</sub>CH<sub>2</sub>-S-CH<sub>2</sub>CH<sub>2</sub>-. A preferred definition of R<sub>2</sub> is, for example, C<sub>4</sub>-C<sub>10</sub>alkylene interrupted by oxygen or sulfur, especially C<sub>4</sub>-C<sub>8</sub>alkylene interrupted by oxygen or sulfur, for example C<sub>4</sub>-C<sub>6</sub>alkylene interrupted by oxygen or sulfur. An especially preferred meaning of R<sub>2</sub> is -CH<sub>2</sub>CH<sub>2</sub>-O-CH<sub>2</sub>CH<sub>2</sub>-O-CH<sub>2</sub>CH<sub>2</sub>- or -CH<sub>2</sub>CH<sub>2</sub>-S-CH<sub>2</sub>CH<sub>2</sub>-.

Alkanetetrayl having 4 to 10 carbon atoms is, for example, 
$$\begin{array}{c} \text{CH}_2- \\ | \\ -\text{CH}_2-\text{C}-\text{CH}_2- \\ | \\ \text{CH}_2- \end{array}$$

(pentaerythryl), 
$$\begin{array}{c} | \quad | \\ -\text{CH}_2-\text{CH}-\text{CH}-\text{CH}_2- \end{array} \quad , \quad \begin{array}{c} | \quad | \\ -\text{CH}_2-\text{CH}_2-\text{CH}-\text{CH}-\text{CH}_2- \end{array} \quad ,$$
  

$$\begin{array}{c} | \quad | \\ -\text{CH}_2-\text{CH}_2-\text{CH}-\text{CH}-\text{CH}_2-\text{CH}_2- \end{array} \quad , \quad \begin{array}{c} | \quad | \\ -\text{CH}_2-\text{CH}_2-\text{CH}-\text{CH}_2-\text{CH}-\text{CH}_2-\text{CH}_2- \end{array} \quad \text{or}$$
  

$$\begin{array}{c} | \quad | \\ -\text{CH}_2-\text{CH}_2-\text{CH}-\text{CH}_2-\text{CH}_2-\text{CH}-\text{CH}_2-\text{CH}_2- \end{array} \quad . \text{ Pentaerythryl is preferred.}$$

Component (d) may also comprise mixtures of different sterically hindered phenols of the formula VI.

Compositions which are of interest include those comprising as component (d) at least one compound of the formula VI in which, if n is 1, R<sub>2</sub> is C<sub>1</sub>-C<sub>20</sub>alkyl.

Preference is given to compositions comprising as component (d) at least one compound of the formula VI in which,

if n is 2,

R<sub>2</sub> is C<sub>2</sub>-C<sub>8</sub>alkylene, C<sub>4</sub>-C<sub>8</sub>alkylene interrupted by oxygen or sulfur; or, if Y is -NH-, R<sub>2</sub> is additionally a direct bond; and,

if n is 4,

R<sub>2</sub> is C<sub>4</sub>-C<sub>8</sub>alkanetetrayl.

Preference is likewise given to compositions comprising as component (d) at least one compound of the formula VI in which

R<sub>1</sub> is methyl or tert-butyl,

n is 1, 2 or 4,

X is  $\text{---CH}_2\text{---CH}_2\text{---}\overset{\text{O}}{\underset{\text{||}}{\text{C}}}\text{---Y---}$  ,  
Y is <sup>oxygen</sup>~~hydrogen~~ or -NH-; and,

if n is 1,

R<sub>2</sub> is C<sub>14</sub>-C<sub>18</sub>alkyl; and,

if n is 2,

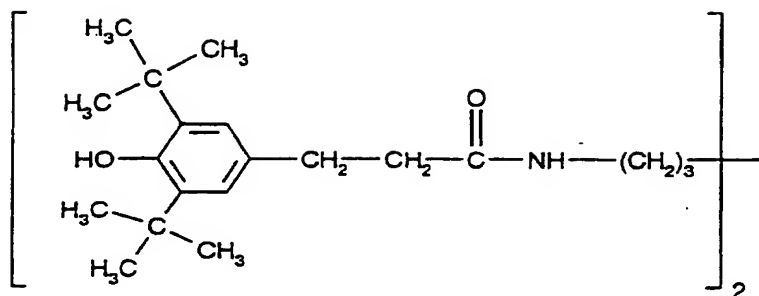
R<sub>2</sub> is C<sub>4</sub>-C<sub>8</sub>alkylene, or is C<sub>4</sub>-C<sub>8</sub>alkylene interrupted by oxygen; and

if n is 4,

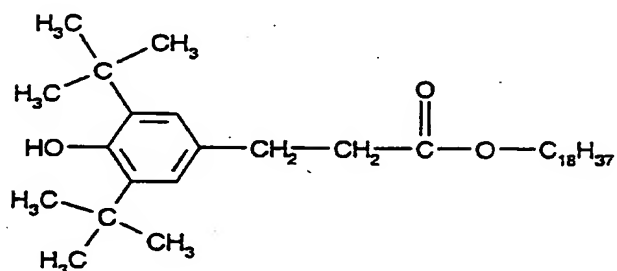
R<sub>2</sub> is C<sub>4</sub>-C<sub>8</sub>alkanetetrayl.

Likewise of interest are compositions comprising as component (d) at least one compound of the formula VI in which the compound of the formula VI is a compound of the formula VIa to VIh

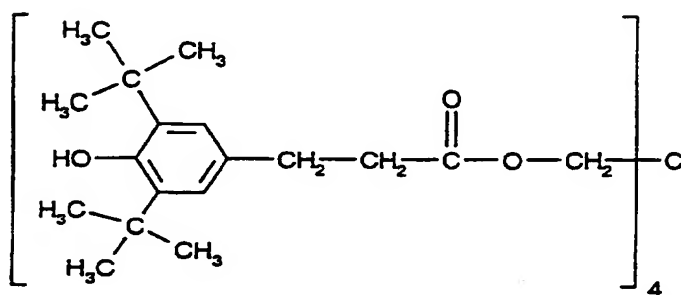




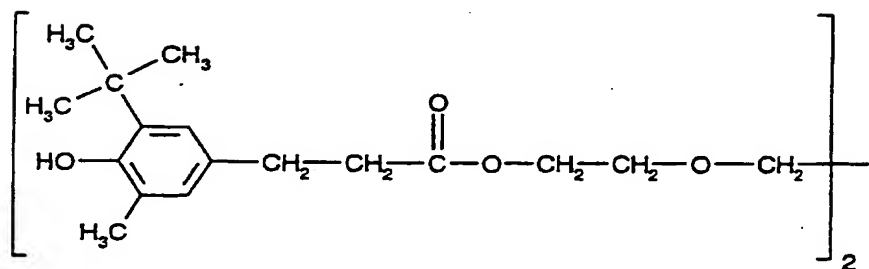
(VIa), Irganox®1098



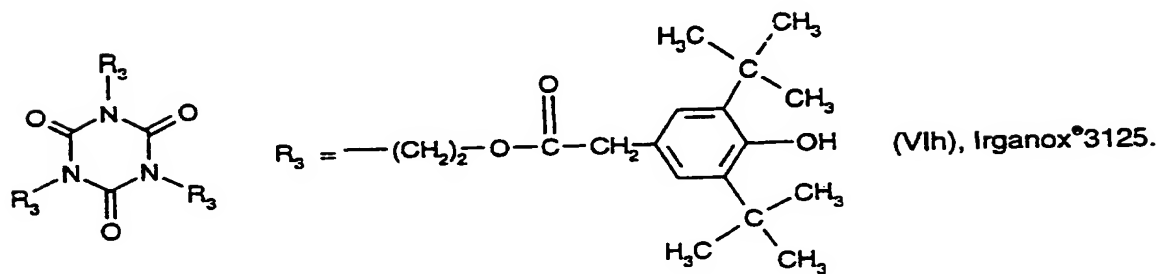
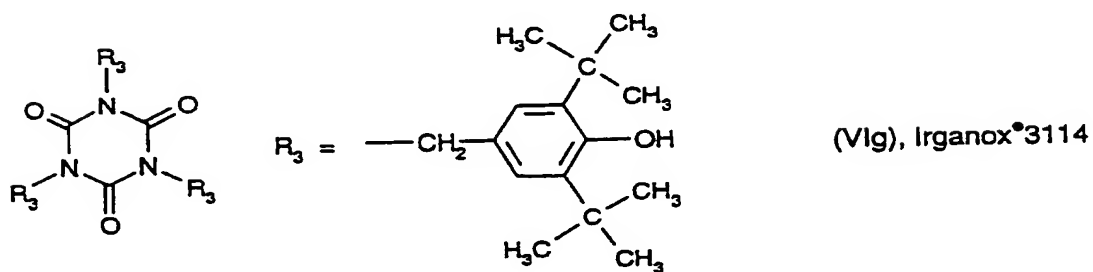
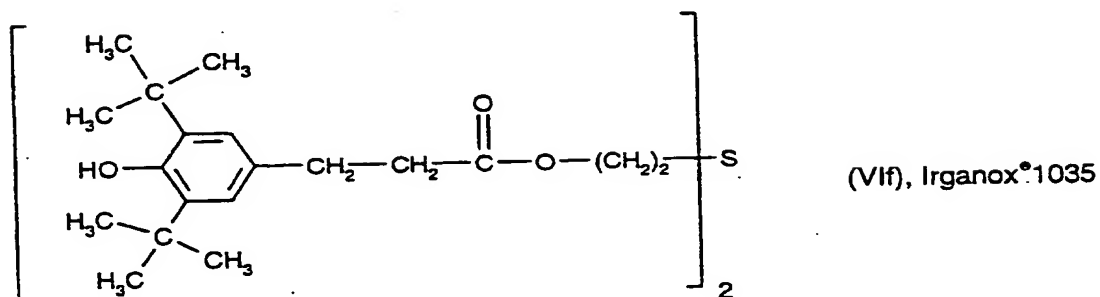
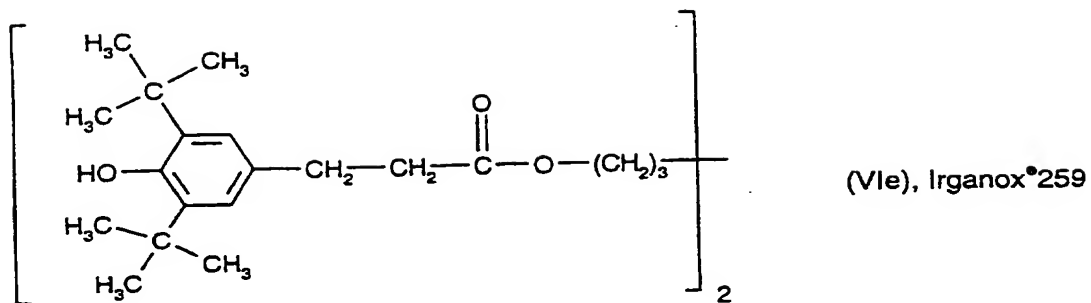
(VIb), Irganox®1076



(VIc), Irganox®1010



(VIId), Irganox®245

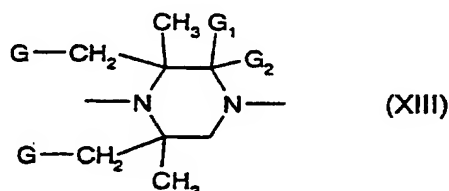
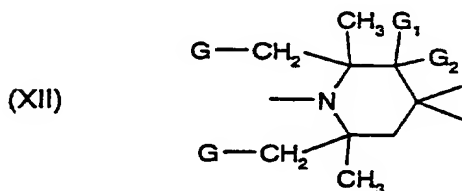


Irganox®1098, Irganox®1076, Irganox®1010, Irganox®245, Irganox®259, Irganox®3114, Irganox®1035 and Irganox®3125 are protected trade names of the company Ciba Spezialitäten-chemie AG.

Preference is given to compositions comprising as component (d) at least one compound of the formula VI in which the compound of the formula VI is a compound of the formula VIa, VIb, VIc or VId, in particular a compound of the formula VIa, VIb or VIc.

Component (d) of the novel composition, and the compounds of the formula VI, are known and in some cases obtainable commercially. Possible preparation processes for the compounds of the formula VI can be found, for example, in the US Patents 3,330,859 or 3,960,928.

Compositions of interest include those comprising as component (e) at least one radical of the formula XII or XIII



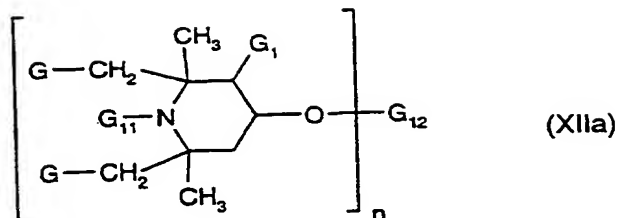
in which

G is hydrogen or methyl, and

G<sub>1</sub> and G<sub>2</sub> are hydrogen, methyl or together are oxygen.

Of particular interest are compositions comprising as component (e) at least one compound from the group of the sterically hindered amines of the class of compounds described under (a') to (g'), which comprise at least one radical of the formula XII or XIII.

(a') Compounds of the formula XIIa



in which n is a number from 1 to 4,

G and G<sub>1</sub> independently of one another are hydrogen or methyl,

G<sub>11</sub> is hydrogen, O<sup>+</sup>, hydroxyl, NO, -CH<sub>2</sub>CN, C<sub>1</sub>-C<sub>18</sub>alkyl, C<sub>3</sub>-C<sub>8</sub>alkenyl, C<sub>3</sub>-C<sub>8</sub>alkynyl,

C<sub>7</sub>-C<sub>12</sub>aralkyl, C<sub>1</sub>-C<sub>18</sub>alkoxy, C<sub>5</sub>-C<sub>8</sub>cycloalkoxy, C<sub>7</sub>-C<sub>9</sub>phenylalkoxy, C<sub>1</sub>-C<sub>8</sub>alkanoyl,

C<sub>3</sub>-C<sub>5</sub>alkenoyl, C<sub>1</sub>-C<sub>18</sub>alkanoyloxy, benzyloxy, glycidyl or a group -CH<sub>2</sub>CH(OH)-Z, where G<sub>11</sub>

is preferably hydrogen, C<sub>1</sub>-C<sub>4</sub>alkyl, allyl, benzyl, acetyl or acryloyl,

Z is hydrogen, methyl or phenyl, and,

if n is 1,

G<sub>12</sub> is hydrogen, C<sub>1</sub>-C<sub>18</sub>alkyl, which is uninterrupted or interrupted by one or more oxygen atoms, or is cyanoethyl, benzyl, glycidyl, a monovalent radical of an aliphatic, cycloaliphatic, araliphatic, unsaturated or aromatic carboxylic acid, carbamic acid or phosphorus-containing acid or a monovalent silyl radical, preferably a radical of an aliphatic carboxylic acid having 2 to 18 carbon atoms, of a cycloaliphatic carboxylic acid having 7 to 15 carbon atoms, of an α,β-unsaturated carboxylic acid having 3 to 5 carbon atoms or of an aromatic carboxylic acid having 7 to 15 carbon atoms, it being possible for the carboxylic acid to be substituted in

each case in the aliphatic, cycloaliphatic or aromatic moiety from 1 to 3 times by -COOZ<sub>12</sub>,

Z<sub>12</sub> is hydrogen, C<sub>1</sub>-C<sub>20</sub>alkyl, C<sub>3</sub>-C<sub>12</sub>alkenyl, C<sub>5</sub>-C<sub>7</sub>cycloalkyl, phenyl or benzyl, and,

if n is 2,

G<sub>12</sub> is C<sub>2</sub>-C<sub>12</sub>alkylene, C<sub>4</sub>-C<sub>12</sub>alkenylene, xylylene, a divalent radical of an aliphatic, cycloaliphatic, araliphatic or aromatic dicarboxylic acid, dicarbamic acid or phosphorus-containing acid or a divalent silyl radical, preferably a radical of an aliphatic dicarboxylic acid having 2 to 36 carbon atoms, of a cycloaliphatic or aromatic dicarboxylic acid having 8 to 14 carbon atoms or of an aliphatic, cycloaliphatic or aromatic dicarbamic acid having 8 to 14 carbon atoms, it being possible for the dicarboxylic acid to be substituted in each case in the aliphatic, cycloaliphatic or aromatic moiety by 1 or 2 groups of -COOZ<sub>12</sub>, and,

if n is 3,

$G_{12}$  is a trivalent radical of an aliphatic, cycloaliphatic or aromatic tricarboxylic acid, which can be substituted in the aliphatic, cycloaliphatic or aromatic moiety by  $-COOZ_{12}$ , of an aromatic tricarbamic acid or of a phosphorus-containing acid, or is a trivalent silyl radical, and,

if  $n$  is 4,

$G_{12}$  is a tetravalent radical of an aliphatic, cycloaliphatic or aromatic tetracarboxylic acid.

The carboxylic acid radicals indicated include in each case radicals of the formula  $(-CO)_nR$ , in which the meaning of  $n$  is indicated above and the meaning of  $R$  is evident from the definition stated.

Any  $C_1$ - $C_{12}$ alkyl substituents are, for example, methyl, ethyl, *n*-propyl, *n*-butyl, *sec*-butyl, *tert*-butyl, *n*-hexyl, *n*-octyl, 2-ethylhexyl, *n*-nonyl, *n*-decyl, *n*-undecyl or *n*-dodecyl.

As  $C_1$ - $C_{18}$ alkyl  $G_{11}$  or  $G_{12}$  can be, for example, the groups indicated above and also, for example, *n*-tridecyl, *n*-tetradecyl, *n*-hexadecyl or *n*-octadecyl.

If  $G_{11}$  is  $C_3$ - $C_8$ alkenyl it can, for example, be 1-propenyl, allyl, methallyl, 2-butenyl, 2-pentenyl, 2-hexenyl, 2-octenyl or 4-*tert*-butyl-2-butenyl.

$G_{11}$  as  $C_3$ - $C_8$ alkynyl is preferably propargyl.

$G_{11}$  as  $C_7$ - $C_{12}$ aralkyl is especially phenethyl and in particular benzyl.

$G_{11}$  as  $C_1$ - $C_8$ alkanoyl is, for example, formyl, propionyl, butyryl, octanoyl, but preferably acetyl and as  $C_3$ - $C_5$ alkenoyl is especially acryloyl.

$G_{12}$  as a monovalent radical of a carboxylic acid is, for example, an acetic, caproic, stearic, acrylic, methacrylic, benzoic or  $\beta$ -(3,5-di-*tert*-butyl-4-hydroxyphenyl)propionic acid radical.

If  $G_{12}$  is a monovalent silyl radical then it is, for example, a radical of the formula  $-(C_jH_{2j})-Si(Z')_2Z''$  in which  $j$  is an integer from the range from 2 to 5 and  $Z'$  and  $Z''$  independently of one another are  $C_1$ - $C_4$ alkyl or  $C_1$ - $C_4$ alkoxy.

If  $G_{12}$  is a divalent radical of a dicarboxylic acid then it is, for example, a malonic, succinic, glutaric, adipic, suberic, sebacic, maleic, itaconic, phthalic, dibutylmalonic, dibenzylmalonic, butyl(3,5-di-tert-butyl-4-hydroxybenzyl)malonic or bicycloheptenedicarboxylic acid radical.

If  $G_{12}$  is a trivalent radical of a tricarboxylic acid then it is, for example, a trimellitic, citric or nitrilotriacetic acid radical.

If  $G_{12}$  is a tetravalent radical of a tetracarboxylic acid then it is, for example, the tetravalent radical of butane-1,2,3,4-tetracarboxylic acid or of pyromellitic acid.

If  $G_{12}$  is a divalent radical of a dicarbamic acid then it is, for example, a hexamethylenedicarbamic or a 2,4-tolylenedicarbamic acid radical.

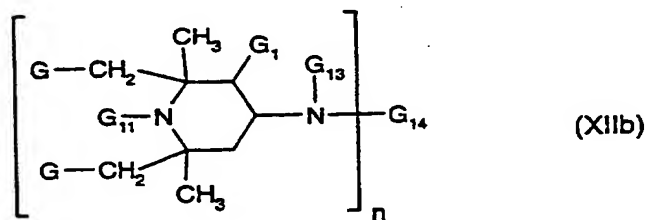
Preference is given to compounds of the formula XIIa in which G is hydrogen,  $G_{11}$  is hydrogen or methyl, n is 2 and  $G_{12}$  is the diacyl radical of an aliphatic dicarboxylic acid having 4 to 12 carbon atoms.

Examples of polyalkylpiperidine compounds of this class are the following compounds:

- 1) 4-Hydroxy-2,2,6,6-tetramethylpiperidine
- 2) 1-Allyl-4-hydroxy-2,2,6,6-tetramethylpiperidine
- 3) 1-Benzyl-4-hydroxy-2,2,6,6-tetramethylpiperidine
- 4) 1-(4-tert-Butyl-2-butenyl)-4-hydroxy-2,2,6,6-tetramethylpiperidine
- 5) 4-Stearoyloxy-2,2,6,6-tetramethylpiperidine
- 6) 1-Ethyl-4-salicyloyloxy-2,2,6,6-tetramethylpiperidine
- 7) 4-Methacryloyloxy-1,2,2,6,6-pentamethylpiperidine
- 8) 1,2,2,6,6-Pentamethylpiperidin-4-yl  $\beta$ -(3,5-di-tert-butyl-4-hydroxyphenyl)propionate
- 9) Di(1-benzyl-2,2,6,6-tetramethylpiperidin-4-yl) maleate
- 10) Di(2,2,6,6-tetramethylpiperidin-4-yl) succinate
- 11) Di(2,2,6,6-tetramethylpiperidin-4-yl) glutarate
- 12) Di(2,2,6,6-tetramethylpiperidin-4-yl) adipate
- 13) Di(2,2,6,6-tetramethylpiperidin-4-yl) sebacate
- 14) Di(1,2,2,6,6-pentamethylpiperidin-4-yl) sebacate
- 15) Di(1,2,3,6-tetramethyl-2,6-diethylpiperidin-4-yl) sebacate

- 16) Di(1-allyl-2,2,6,6-tetramethylpiperidin-4-yl) phthalate
- 17) 1-Hydroxy-4-β-cyanoethoxy-2,2,6,6-tetramethylpiperidine
- 18) 1-Acetyl-2,2,6,6-tetramethylpiperidin-4-yl acetate
- 19) Tri(2,2,6,6-tetramethylpiperidin-4-yl) trimellitate
- 20) 1-Acryloyl-4-benzyloxy-2,2,6,6-tetramethylpiperidine
- 21) Di(2,2,6,6-tetramethylpiperidin-4-yl) diethylmalonate
- 22) Di(1,2,2,6,6-pentamethylpiperidin-4-yl) dibutylmalonate
- 23) Di(1,2,2,6,6-pentamethylpiperidin-4-yl) butyl(3,5-di-tert-butyl-4-hydroxybenzyl)malonate
- 24) Di(1-octyloxy-2,2,6,6-tetramethylpiperidin-4-yl) sebacate
- 25) Di(1-cyclohexyloxy-2,2,6,6-tetramethylpiperidin-4-yl) sebacate
- 26) Hexane-1',6'-bis(4-carbamoyloxy-1-n-butyl-2,2,6,6-tetramethylpiperidine)
- 27) Toluene-2',4'-bis(4-carbamoyloxy-1-n-propyl-2,2,6,6-tetramethylpiperidine)
- 28) Dimethylbis(2,2,6,6-tetramethylpiperidin-4-oxy)silane
- 29) Phenyltris(2,2,6,6-tetramethylpiperidin-4-oxy)silane
- 30) Tris(1-propyl-2,2,6,6-tetramethylpiperidin-4-yl) phosphite
- 31) Tris(1-propyl-2,2,6,6-tetramethylpiperidin-4-yl) phosphate
- 32) Phenyl [bis(1,2,2,6,6-pentamethylpiperidin-4-yl)]phosphonate
- 33) 4-Hydroxy-1,2,2,6,6-pentamethylpiperidine
- 34) 4-Hydroxy-N-hydroxyethyl-2,2,6,6-tetramethylpiperidine
- 35) 4-Hydroxy-N-(2-hydroxypropyl)-2,2,6,6-tetramethylpiperidine
- 36) 1-Glycidyl-4-hydroxy-2,2,6,6-tetramethylpiperidine.

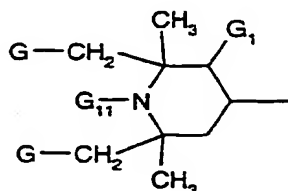
(b') Compounds of the formula XIIb



in which n is the number 1 or 2,

G, G<sub>1</sub> and G<sub>11</sub> are as defined under (a'),

$G_{13}$  is hydrogen,  $C_1$ - $C_{12}$ alkyl,  $C_2$ - $C_5$ hydroxyalkyl,  $C_5$ - $C_7$ cycloalkyl,  $C_7$ - $C_8$ aralkyl,  $C_2$ - $C_{18}$ alkanoyl,  $C_3$ - $C_5$ alkenoyl, benzoyl or a group of the formula



and,

if  $n$  is 1,

$G_{14}$  is hydrogen,  $C_1$ - $C_{18}$ alkyl,  $C_3$ - $C_8$ alkenyl,  $C_5$ - $C_7$ cycloalkyl, or  $C_1$ - $C_4$ alkyl substituted by a hydroxyl, cyano, alkoxy carbonyl or carbamide group; glycidyl, a group of the formula  $-CH_2-CH(OH)-Z$  or of the formula  $-CONH-Z$ , in which  $Z$  is hydrogen, methyl or phenyl, and if  $n$  is 2,

$G_{14}$  is  $C_2$ - $C_{12}$ alkylene,  $C_6$ - $C_{12}$ arylene, xylylene, a  $-CH_2-CH(OH)-CH_2-$  group or a group  $-CH_2-CH(OH)-CH_2-O-D-O-$  in which  $D$  is  $C_2$ - $C_{10}$ alkylene,  $C_6$ - $C_{15}$ arylene,  $C_6$ - $C_{12}$ cycloalkylene, or, provided that  $G_{13}$  is not alkanoyl, alkenoyl or benzoyl,  $G_{14}$  can alternatively be 1-oxo- $C_2$ - $C_{12}$ alkylene, a divalent radical of an aliphatic, cycloaliphatic or aromatic dicarboxylic acid or dicarbamic acid or else can be the group  $-CO-$ , or,

if  $n$  is 1,

$G_{13}$  and  $G_{14}$  together can be the divalent radical of an aliphatic, cycloaliphatic or aromatic 1,2- or 1,3-dicarboxylic acid.

Any  $C_1$ - $C_{12}$  or  $C_1$ - $C_{18}$ alkyl substituents are as already defined under (a').

Any  $C_5$ - $C_7$ cycloalkyl substituents are, in particular, cyclohexyl.

$G_{13}$  as  $C_7$ - $C_8$ aralkyl is in particular phenylethyl or especially benzyl. As  $C_2$ - $C_5$ hydroxyalkyl  $G_{13}$  is, in particular, 2-hydroxyethyl or 2-hydroxypropyl.

$G_{13}$  as  $C_2$ - $C_{18}$ alkanoyl is, for example, propionyl, butyryl, octanoyl, dodecanoyl, hexadecanoyl, octadecanoyl, but preferably acetyl, and as  $C_3$ - $C_5$ alkenoyl is especially acryloyl.



If  $G_{14}$  is  $C_2$ - $C_8$ alkenyl it is, for example, allyl, methallyl, 2-butenyl, 2-pentenyl, 2-hexenyl or 2-octenyl.

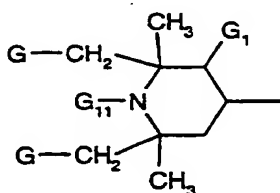
$G_{14}$  as  $C_1$ - $C_4$ alkyl substituted by a hydroxyl, cyano, alkoxycarbonyl or carbamide group can, for example, be 2-hydroxyethyl, 2-hydroxypropyl, 2-cyanoethyl, methoxycarbonylmethyl, 2-ethoxycarbonyl ethyl, 2-aminocarbonylpropyl or 2-(dimethylaminocarbonyl)ethyl.

Any  $C_2$ - $C_{12}$ alkylene substituents are, for example, ethylene, propylene, 2,2-dimethylpropylene, tetramethylene, hexamethylene, octamethylene, decamethylene or dodecamethylene.

Any  $C_6$ - $C_{15}$ arylene substituents are, for example, o-, m- or p-phenylene, 1,4-naphthylene or 4,4'-diphenylene.

As  $C_6$ - $C_{12}$ cycloalkylene particular mention may be made of cyclohexylene.

Preference is given to compounds of the formula Ib in which n is 1 or 2, G is hydrogen,  $G_{11}$  is hydrogen or methyl,  $G_{13}$  is hydrogen,  $C_1$ - $C_{12}$ alkyl or a group of the formula

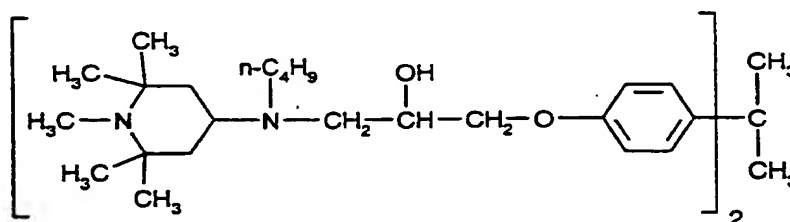


and  $G_{14}$ , if  $n=1$ , is hydrogen or  $C_1$ - $C_{12}$ alkyl and, if  $n=2$ , is  $C_2$ - $C_8$ alkylene or 1-oxo- $C_2$ - $C_8$ alkylene.

Examples of polyalkylpiperidine compounds of this class are the following compounds:

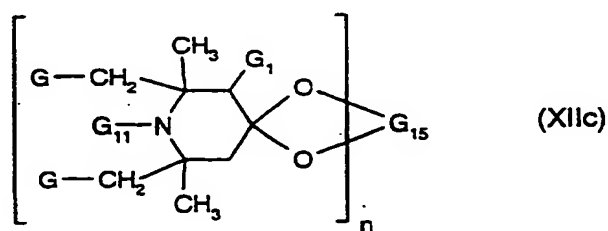
- 37) N,N'-Bis(2,2,6,6-tetramethylpiperidin-4-yl)hexamethylene-1,6-diamine
- 38) N,N'-Bis(2,2,6,6-tetramethylpiperidin-4-yl)hexamethylene-1,6-diacetamide
- 39) Bis(2,2,6,6-tetramethylpiperidin-4-yl)amine
- 40) 4-Benzoylamino-2,2,6,6-tetramethylpiperidine
- 41) N,N'-Bis(2,2,6,6-tetramethylpiperidin-4-yl)-N,N'-dibutyladipamide

- 42) N,N'-Bis(2,2,6,6-tetramethylpiperidin-4-yl)-N,N'-dicyclohexyl-2-hydroxypropylene-1,3-diamine  
 43) N,N'-Bis(2,2,6,6-tetramethylpiperidin-4-yl)-p-xylylenediamine  
 44) N,N'-Bis(2,2,6,6-tetramethylpiperidin-4-yl)succinamide  
 45) Di(2,2,6,6-tetramethylpiperidin-4-yl) N-(2,2,6,6-tetramethylpiperidin-4-yl)-β-amino-dipropionate  
 46) The compound of the formula



- 47) 4-(Bis-2-hydroxyethylamino)-1,2,2,6,6-pentamethylpiperidine  
 48) 4-(3-Methyl-4-hydroxy-5-tert-butylbenzamido)-2,2,6,6-tetramethylpiperidine  
 49) 4-Methacrylamido-1,2,2,6,6-pentamethylpiperidine

(c') Compounds of the formula XIc



in which n is the number 1 or 2, G, G<sub>1</sub> and G<sub>11</sub> are as defined under (a'), and,  
 if n is 1,  
 G<sub>15</sub> is C<sub>2</sub>-C<sub>8</sub>alkylene or C<sub>2</sub>-C<sub>8</sub>hydroxyalkylene or C<sub>4</sub>-C<sub>22</sub>acyloxyalkylene, and,  
 if n is 2,  
 G<sub>15</sub> is the group (-CH<sub>2</sub>)<sub>2</sub>C(CH<sub>2</sub>)<sub>2</sub>.

If  $G_{15}$  is  $C_2$ - $C_8$ alkylene or  $C_2$ - $C_8$ hydroxyalkylene then it is, for example, ethylene, 1-methyl-ethylene, propylene, 2-ethylpropylene or 2-ethyl-2-hydroxymethylpropylene.

$G_{15}$  as  $C_4$ - $C_{22}$ acyloxyalkylene is, for example, 2-ethyl-2-acetoxymethylpropylene.

Examples of polyalkylpiperidine compounds of this class are the following compounds:

50) 9-Aza-8,8,10,10-tetramethyl-1,5-dioxaspiro[5.5]undecane

51) 9-Aza-8,8,10,10-tetramethyl-3-ethyl-1,5-dioxaspiro[5.5]undecane

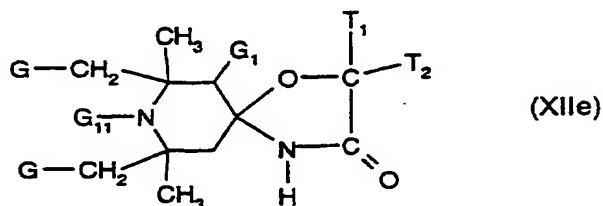
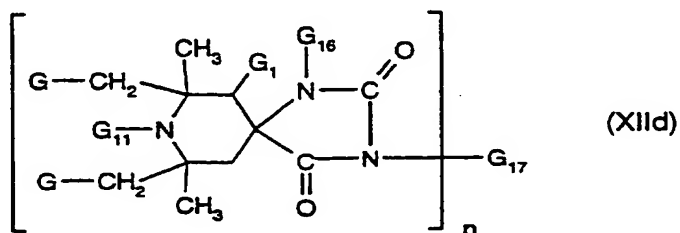
52) 8-Aza-2,7,7,8,9,9-hexamethyl-1,4-dioxaspiro[4.5]decane

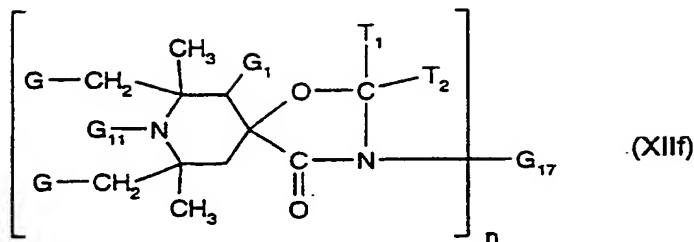
53) 9-Aza-3-hydroxymethyl-3-ethyl-8,8,9,10,10-pentamethyl-1,5-dioxaspiro[5.5]undecane

54) 9-Aza-3-ethyl-3-acetoxymethyl-9-acetyl-8,8,10,10-tetramethyl-1,5-dioxaspiro[5.5]undecane

55) 2,2,6,6-Tetramethylpiperidine-4-spiro-2'-(1',3'-dioxane)-5'-spiro-5''-(1'',3''-dioxane)-2''-spiro-4'''-(2'',2'',6'',6''-tetramethylpiperidine).

(d') Compounds of the formulae XIId, XIle and XIIf, with compounds of the formula XIIf being preferred





in which  $n$  is the number 1 or 2,  $G$ ,  $G_1$  and  $G_{11}$  are as defined under (a'),

$G_{16}$  is hydrogen,  $C_1$ - $C_{12}$ alkyl, allyl, benzyl, glycidyl or  $C_2$ - $C_6$ alkoxyalkyl, and,

if  $n$  is 1,

$G_{17}$  is hydrogen,  $C_1$ - $C_{12}$ alkyl,  $C_3$ - $C_5$ alkenyl,  $C_7$ - $C_9$ aralkyl,  $C_5$ - $C_7$ cycloalkyl,  $C_2$ - $C_4$ hydroxyalkyl,  $C_2$ - $C_6$ alkoxyalkyl,  $C_6$ - $C_{10}$ aryl, glycidyl or a group of the formula

$-(CH_2)_p-COO-Q$  or of the formula  $-(CH_2)_p-O-CO-Q$  in which  $p$  is 1 or 2 and  $Q$  is  $C_1$ - $C_4$ alkyl or phenyl, and,

if  $n$  is 2,

$G_{17}$  is  $C_2$ - $C_{12}$ alkylene,  $C_4$ - $C_{12}$ alkenylene,  $C_6$ - $C_{12}$ arylene, a group

$-CH_2-CH(OH)-CH_2-O-D-O-CH_2-CH(OH)-CH_2-$  in which  $D$  is  $C_2$ - $C_{10}$ alkylene,  $C_6$ - $C_{15}$ arylene,  $C_6$ - $C_{12}$ cycloalkylene, or a group  $-CH_2CH(OZ')CH_2-(OCH_2-CH(OZ')CH_2)_2-$  in which  $Z'$  is hydrogen,  $C_1$ - $C_{18}$ alkyl, allyl, benzyl,  $C_2$ - $C_{12}$ alkanoyl or benzoyl,

$T_1$  and  $T_2$  independently of one another are hydrogen,  $C_1$ - $C_{18}$ alkyl or unsubstituted or halo- or  $C_1$ - $C_4$ alkyl-substituted  $C_6$ - $C_{10}$ aryl or  $C_7$ - $C_9$ aralkyl or  $T_1$  and  $T_2$ , together with the carbon atom to which they are attached, form a  $C_5$ - $C_{14}$ cycloalkane ring.

Any  $C_1$ - $C_{12}$ alkyl substituents are, for example, methyl, ethyl, *n*-propyl, *n*-butyl, *sec*-butyl, *tert*-butyl, *n*-hexyl, *n*-octyl, 2-ethylhexyl, *n*-nonyl, *n*-decyl, *n*-undecyl or *n*-dodecyl.

Any substituents in the meaning of  $C_1$ - $C_{18}$ alkyl can, for example, be the groups listed above and also, for example, *n*-tridecyl, *n*-tetradecyl, *n*-hexadecyl or *n*-octadecyl.

Any  $C_2$ - $C_6$ alkoxyalkyl substituents are, for example, methoxymethyl, ethoxymethyl, propoxymethyl, *tert*-butoxymethyl, ethoxyethyl, ethoxypropyl, *n*-butoxyethyl, *tert*-butoxyethyl, isopropoxyethyl or propoxypropyl.

If  $G_{17}$  is  $C_3$ - $C_5$ alkenyl then it is, for example, 1-propenyl, allyl, methallyl, 2-butenyl or 2-pentenyl.

$G_{17}$ ,  $T_1$  and  $T_2$  as  $C_7$ - $C_9$ aralkyl are, in particular, phenethyl or especially benzyl. If  $T_1$  and  $T_2$  together with the carbon atom form a cycloalkane ring then this can, for example, be a cyclopentane, cyclohexane, cyclooctane or cyclododecane ring.

If  $G_{17}$  is  $C_2$ - $C_4$ hydroxyalkyl then it is, for example, 2-hydroxyethyl, 2-hydroxypropyl, 2-hydroxybutyl or 4-hydroxybutyl.

$G_{17}$ ,  $T_1$  and  $T_2$  as  $C_6$ - $C_{10}$ aryl are, in particular, phenyl,  $\alpha$ - or  $\beta$ -naphthyl which are unsubstituted or substituted by halogen or  $C_1$ - $C_4$ alkyl.

If  $G_{17}$  is  $C_2$ - $C_{12}$ alkylene then it is, for example, ethylene, propylene, 2,2-dimethylpropylene, tetramethylene, hexamethylene, octamethylene, decamethylene or dodecamethylene.

$G_{17}$  as  $C_4$ - $C_{12}$ alkenylene is, in particular, 2-butenylene, 2-pentenylene or 3-hexenylene.

If  $G_{17}$  is  $C_6$ - $C_{12}$ arylene then it is, for example, o-, m- or p-phenylene, 1,4-naphthylene or 4,4'-diphenylene.

If  $Z'$  is  $C_2$ - $C_{12}$ alkanoyl then it is, for example, propionyl, butyryl, octanoyl, dodecanoyl, but preferably acetyl.

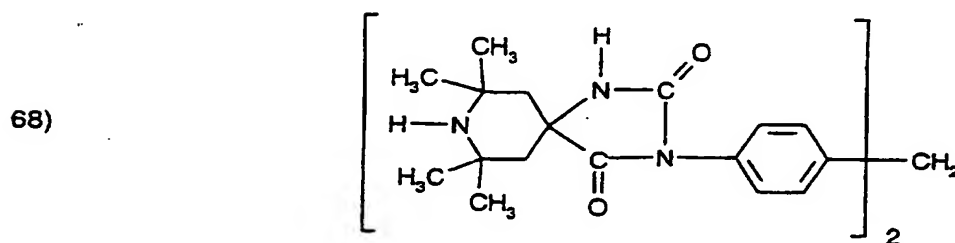
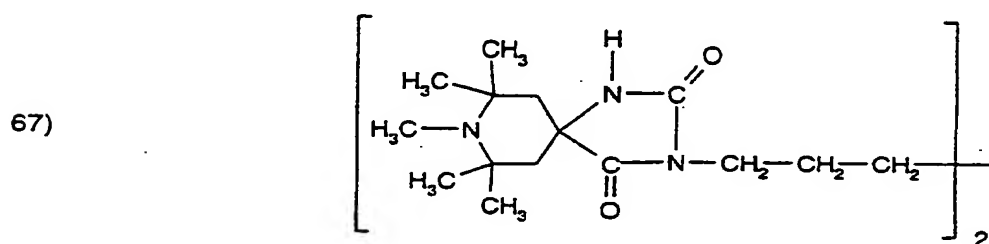
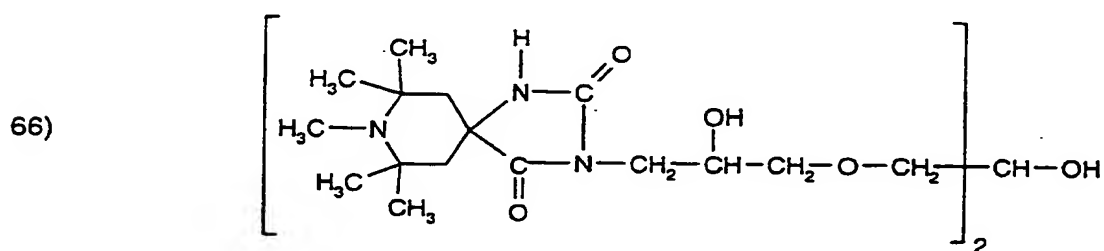
$D$  as  $C_2$ - $C_{10}$ alkylene,  $C_6$ - $C_{15}$ arylene or  $C_6$ - $C_{12}$ cycloalkylene is as defined under (b').

Examples of polyalkylpiperidine compounds of this class are the following compounds:

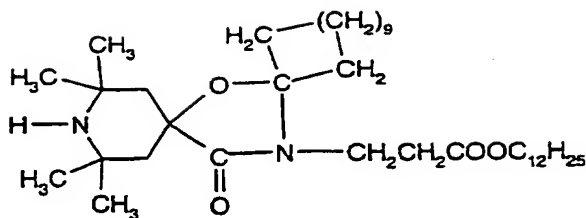
- 56) 3-Benzyl-1,3,8-triaza-7,7,9,9-tetramethylspiro[4.5]decane-2,4-dione
- 57) 3-n-Octyl-1,3,8-triaza-7,7,9,9-tetramethylspiro[4.5]decane-2,4-dione
- 58) 3-Allyl-1,3,8-triaza-1,7,7,9,9-pentamethylspiro[4.5]decane-2,4-dione
- 59) 3-Glycidyl-1,3,8-triaza-7,7,8,9,9-pentamethylspiro[4.5]decane-2,4-dione
- 60) 1,3,7,7,8,9,9-Heptamethyl-1,3,8-triazaspiro[4.5]decane-2,4-dione
- 61) 2-Isopropyl-7,7,9,9-tetramethyl-1-oxa-3,8-diaza-4-oxospiro[4.5]decane

- 62) 2,2-Dibutyl-7,7,9,9-tetramethyl-1-oxa-3,8-diaza-4-oxospiro[4.5]decane  
 63) 2,2,4,4-Tetramethyl-7-oxa-3,20-diaza-21-oxodispiro[5.1.11.2]heneicosane  
 64) 2-Butyl-7,7,9,9-tetramethyl-1-oxa-4,8-diaza-3-oxospiro[4.5]decane and preferably:  
 65) 8-Acetyl-3-dodecyl-1,3,8-triaza-7,7,9,9-tetramethylspiro[4.5]decane-2,4-dione

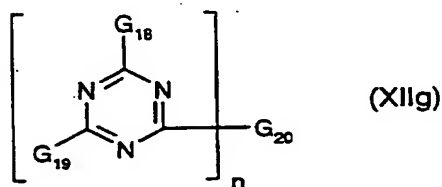
or the compounds of the following formulae:



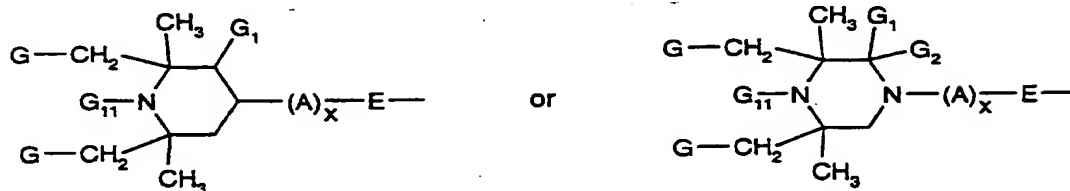
69)



(e') Compounds of the formula XIIg, which in turn are preferred



in which n is the number 1 or 2 and  $G_{18}$  is a group of one of the formulae



in which G and  $G_{11}$  are as defined under (a'),

$G_1$  and  $G_2$  are hydrogen, methyl or together are a substituent =O,

E is -O- or  $-NG_{13}-$ ,

A is  $C_2-C_6$ alkylene or  $-(CH_2)_3-O-$ ,

x is the number 0 or 1,

$G_{13}$  is hydrogen,  $C_1-C_{12}$ alkyl,  $C_2-C_5$ hydroxyalkyl or  $C_5-C_7$ cycloalkyl,

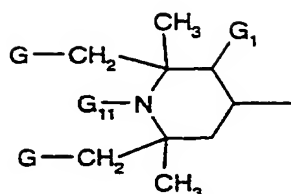
$G_{19}$  is the same as  $G_{18}$  or is one of the group  $-NG_{21}G_{22}$ ,  $-OG_{23}$ ,  $-NHCH_2OG_{23}$  or

$-N(CH_2OG_{23})_2$ ,

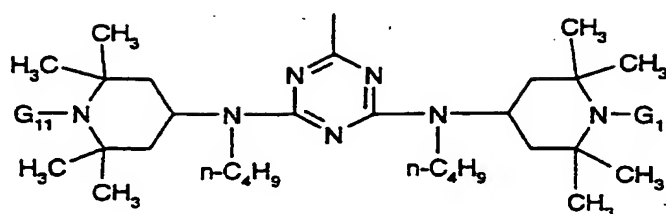
$G_{20}$ , if  $n = 1$ , is the same as  $G_{18}$  or  $G_{19}$  and, if  $n = 2$ ,  $G_{20}$  is a group  $-E-B-E-$ , in which B is

$C_2-C_8$ alkylene or  $C_2-C_8$ alkylene which is interrupted by 1 or 2 groups  $-N(G_{21})-$ ,

$G_{21}$  is  $C_1-C_{12}$ alkyl, cyclohexyl, benzyl or  $C_1-C_4$ hydroxyalkyl or a group of the formula



or a group of the formula



$G_{22}$  is  $C_1$ - $C_{12}$ alkyl, cyclohexyl, benzyl or  $C_1$ - $C_4$ hydroxyalkyl, or  $G_{21}$  and  $G_{22}$  together are  $C_4$ - $C_5$ alkylene or  $C_4$ - $C_5$ oxaalkylene, for example  $-\text{CH}_2\text{CH}_2\text{OCH}_2\text{CH}_2-$  or a group of the formula  $-\text{CH}_2\text{CH}_2\text{N}(G_{11})\text{CH}_2\text{CH}_2-$ , and

$G_{23}$  is hydrogen,  $C_1$ - $C_{12}$ alkyl or phenyl.

Any  $C_1$ - $C_{12}$ alkyl substituents are, for example, methyl, ethyl, n-propyl, n-butyl, sec-butyl, tert-butyl, n-hexyl, n-octyl, 2-ethylhexyl, n-nonyl, n-decyl, n-undecyl or n-dodecyl.

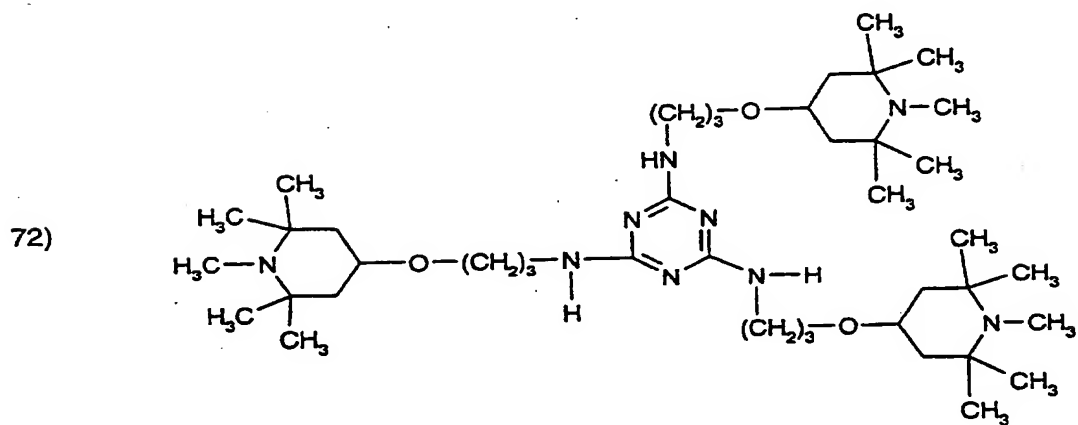
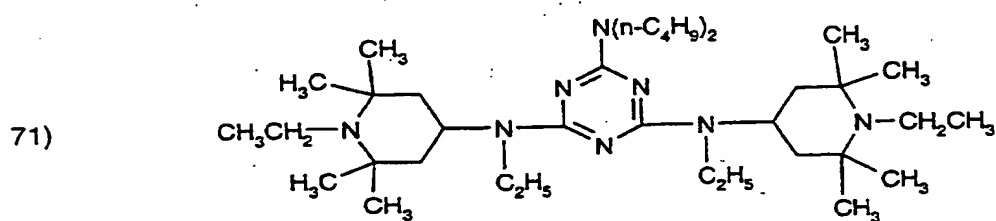
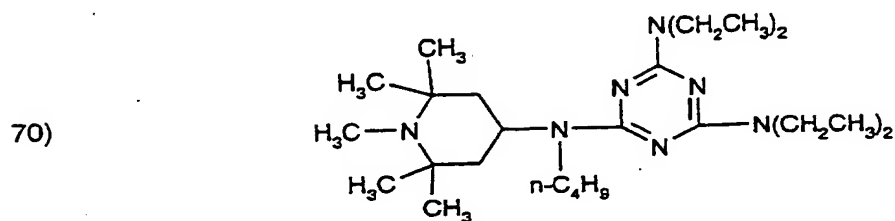
Any  $C_2$ - $C_5$ hydroxyalkyl substituents are, for example, 2-hydroxyethyl, 2-hydroxypropyl, 3-hydroxypropyl, 2-hydroxybutyl or 4-hydroxybutyl.

If A is  $C_2$ - $C_6$ alkylene then it is, for example, ethylene, propylene, 2,2-dimethylpropylene, tetramethylene or hexamethylene.

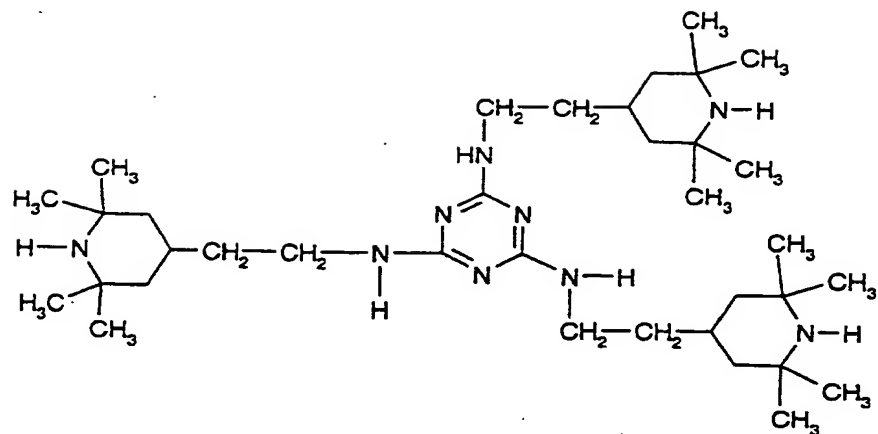
If  $G_{21}$  and  $G_{22}$  together are  $C_4$ - $C_5$ alkylene or oxaalkylene then this is, for example, tetramethylene, pentamethylene or 3-oxapentamethylene.



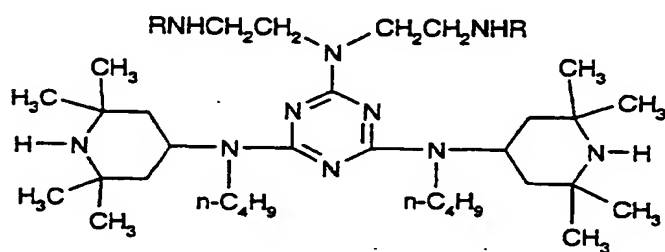
Examples of polyalkylpiperidine compounds of this class are the compounds of the following formulae:



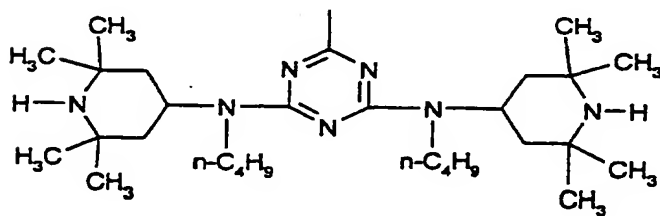
73)



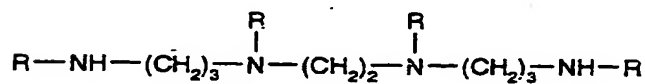
74)



R =

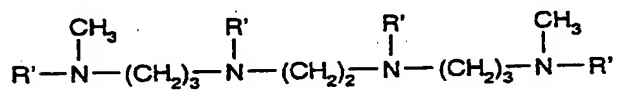
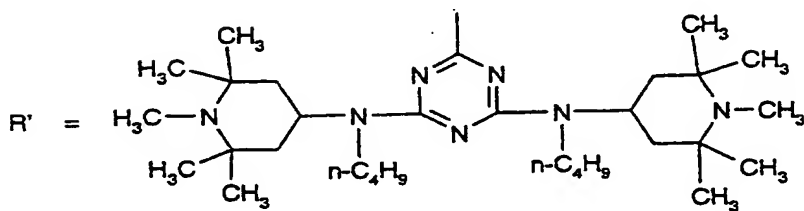


75)

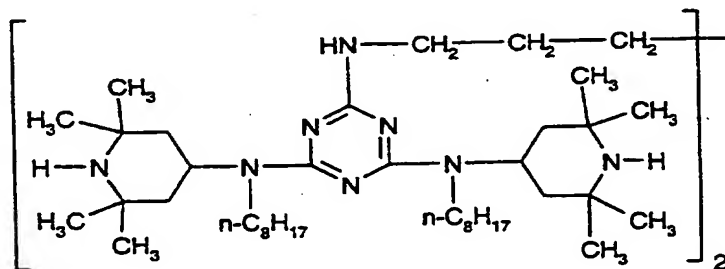


R has the same meaning as in compound 74.

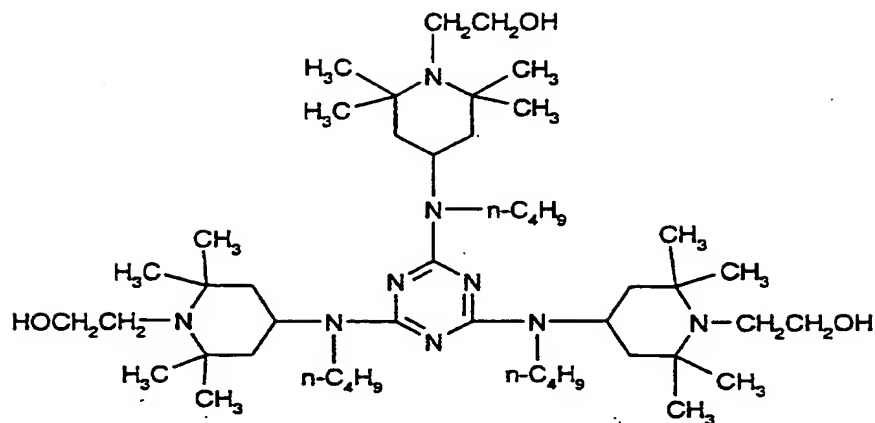
77)



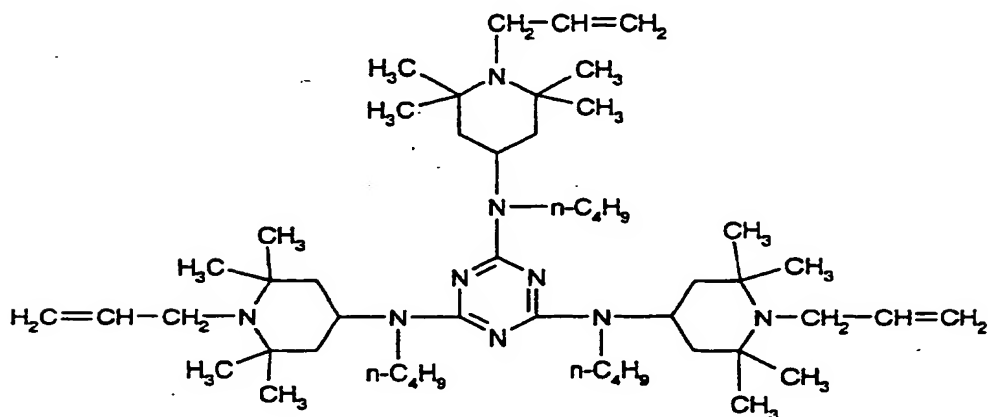
78)



79)

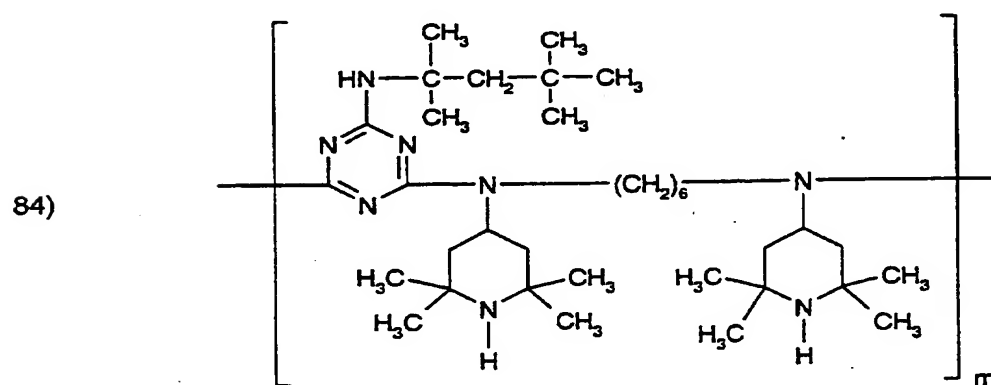
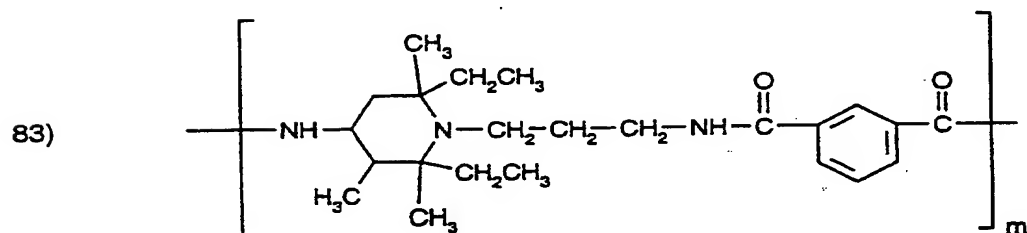
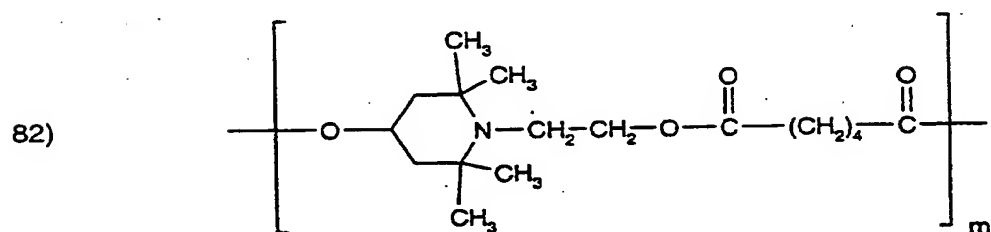
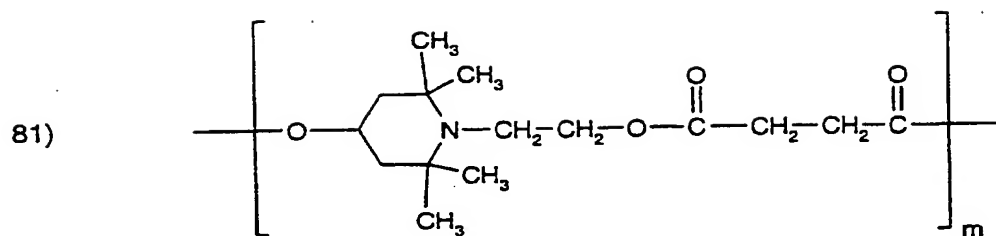


80)

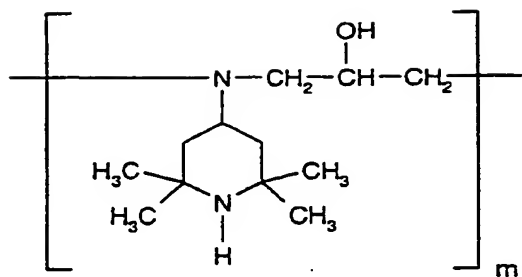


(f') Oligomeric or polymeric compounds whose structural repeating unit contains a 2,2,6,6-tetraalkylpiperidine radical, especially polyesters, polyethers, polyamides, polyamines, polyurethanes, polyureas, polyaminotriazines, poly(meth)acrylates, poly(meth)acrylamides and copolymers thereof which include such radicals.

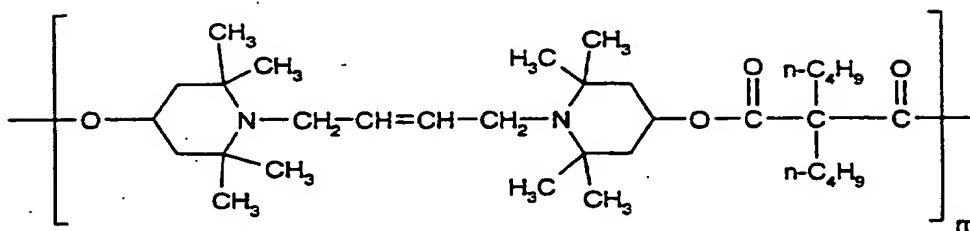
Examples of 2,2,6,6-polyalkylpiperidine compounds of this class are the compounds of the following formulae, in which  $m$  is a number from 2 to about 200.



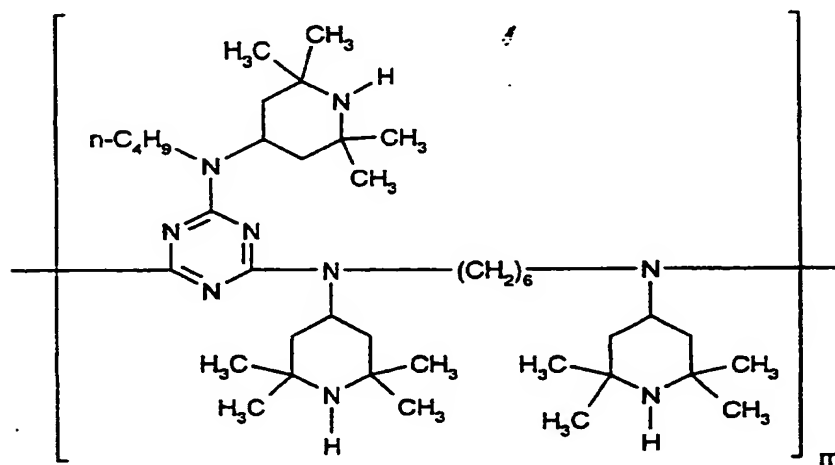
85)

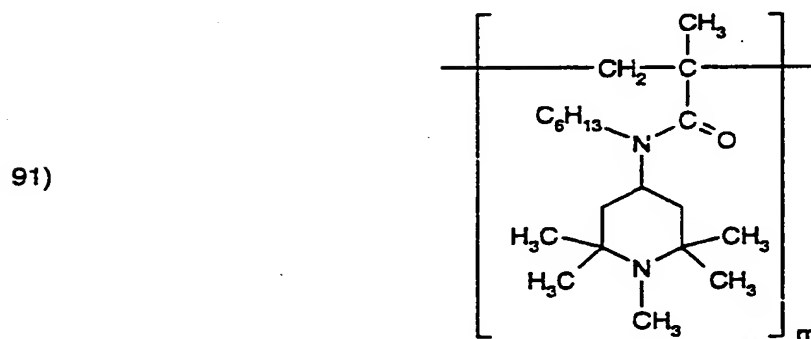
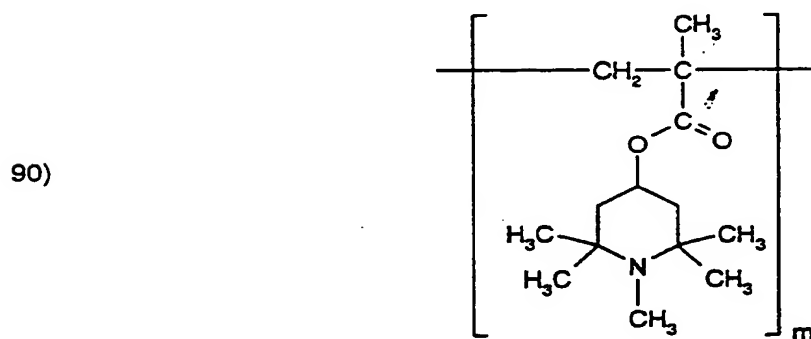
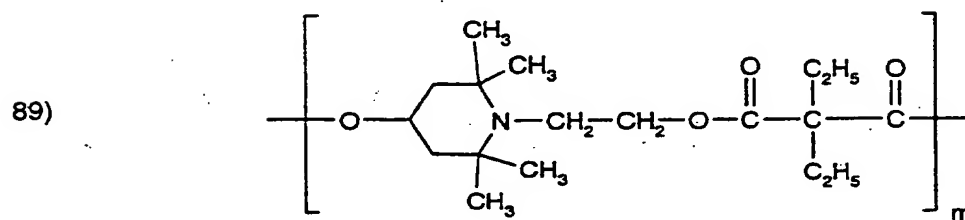
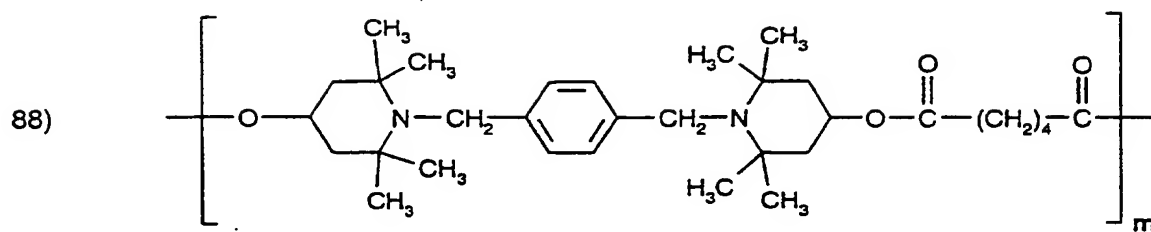


86)

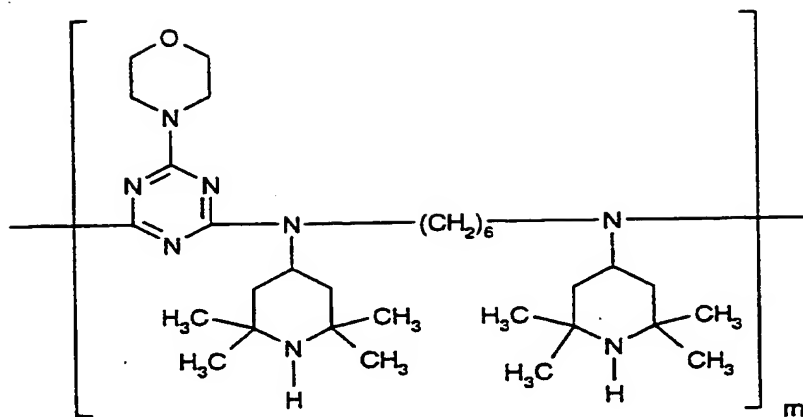


87)

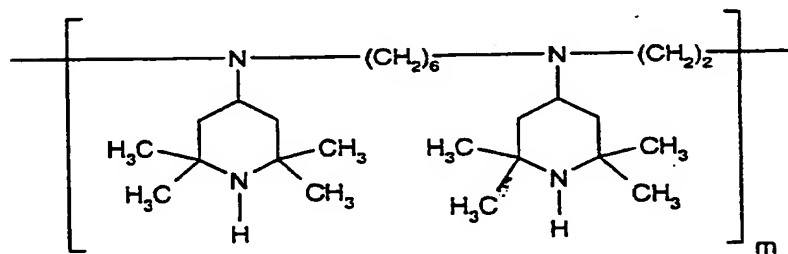




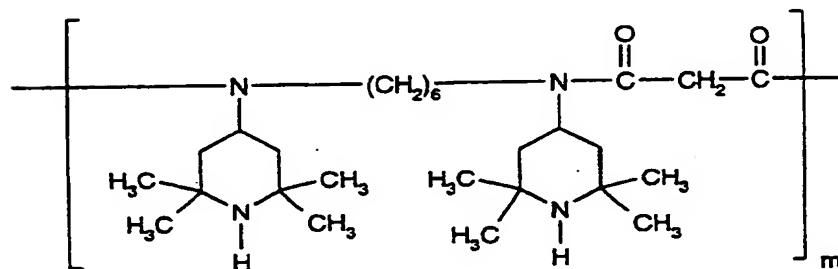
92)



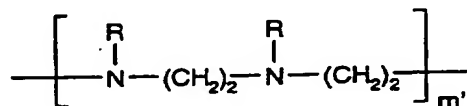
93)



94)

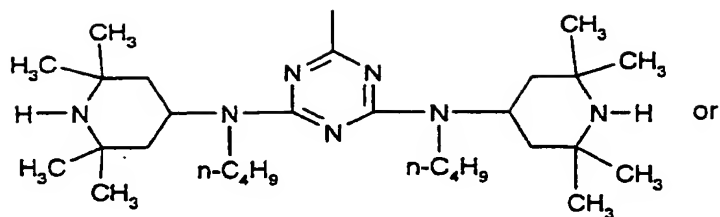


95)

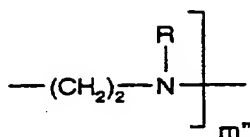




in which R is a radical of the formula



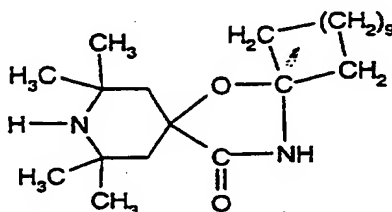
is a chain branch



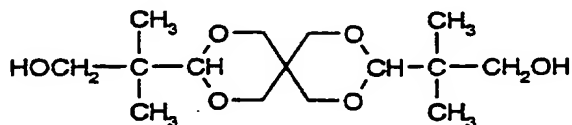
,  $m'$  and  $m''$  are each an integer from the range from

0 to 200, with the proviso that  $m' + m'' = m$ .

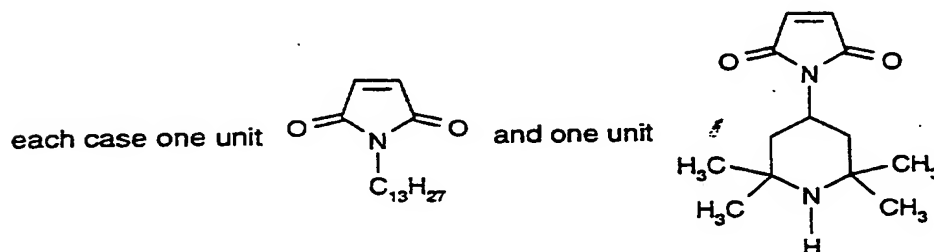
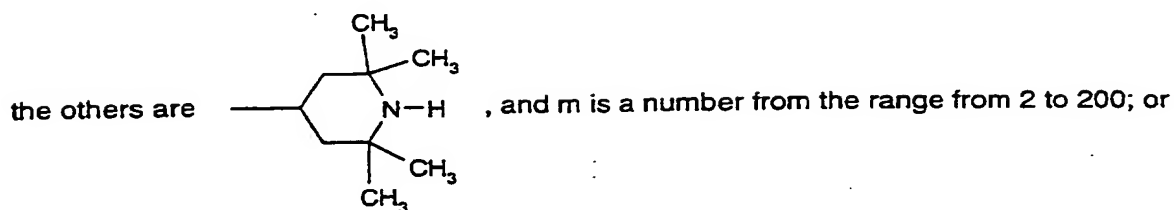
Further examples of polymeric compounds are reaction products of compounds of the formula



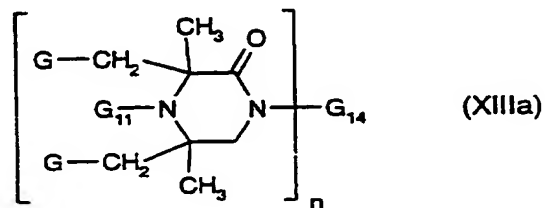
with epichlorohydrin; polyesters of butane-1,2,3,4-tetracarboxylic acid with a bifunctional alcohol of the formula



whose carboxyl side chains originating from the tetracarboxylic acid are esterified with 2,2,6,6-tetramethyl-4-hydroxypiperidine; compounds of the formula

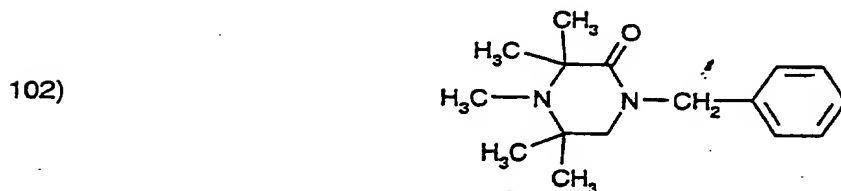
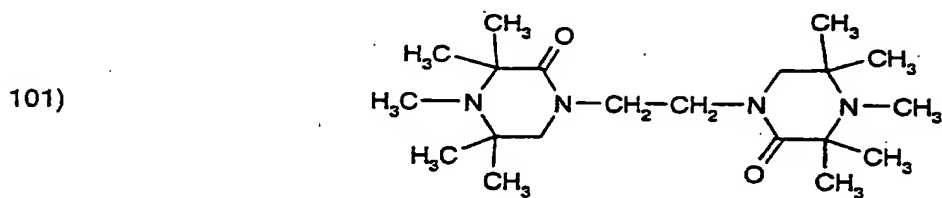
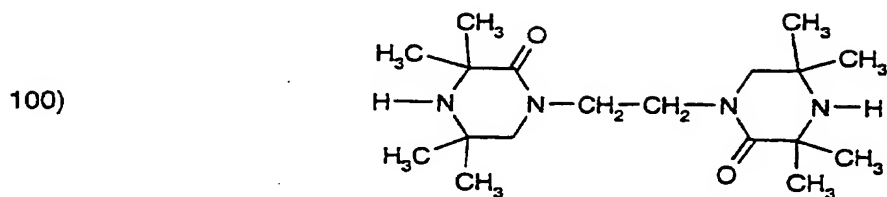


(g') Compounds of the formula XIIIa

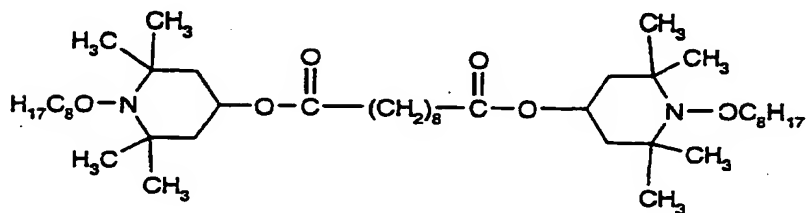


in which n is a number 1 or 2 and in which G and  $\text{G}_{11}$  are as defined under (a') and  $\text{G}_{14}$  is as defined under (b'), the meanings  $-\text{CONH-Z}$  and  $-\text{CH}_2\text{-CH(OH)-CH}_2\text{-O-D-O-}$  being excluded for  $\text{G}_{14}$ .

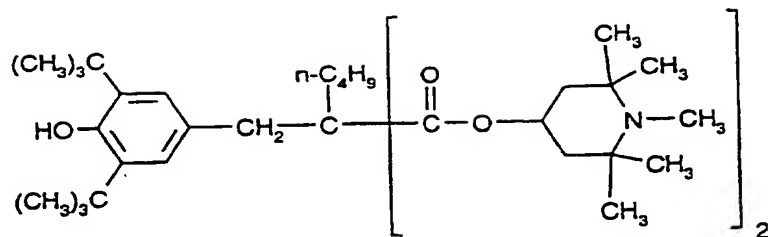
Examples of such compounds are:



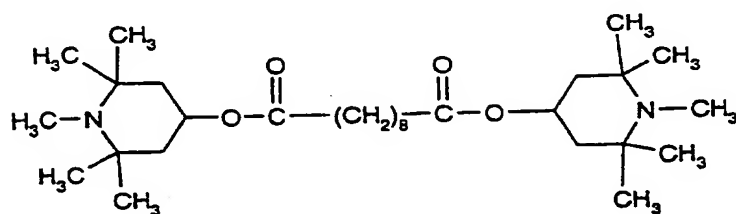
Of particular interest are compositions comprising as component (e) at least one compound of the formula H1, H2, H3, H4, H5, H6 and H7



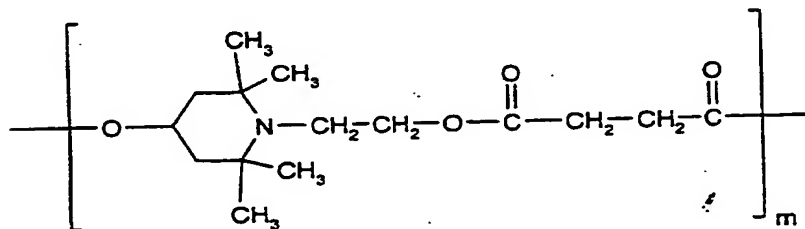
(H1) Tinuvin®123



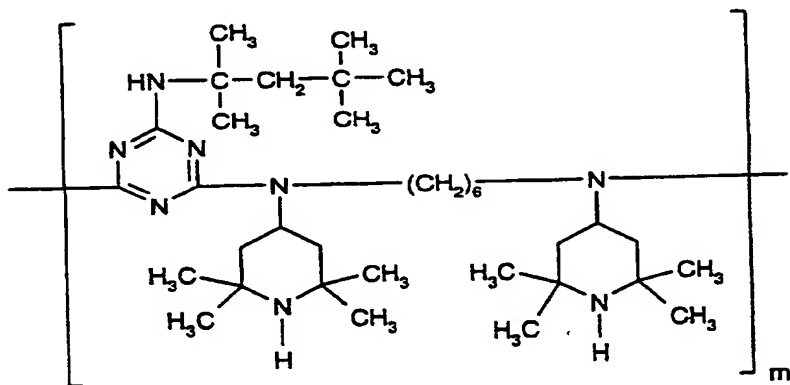
(H2) Tinuvin®144



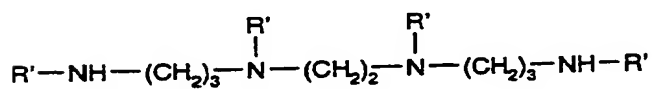
(H3) Tinuvin®292



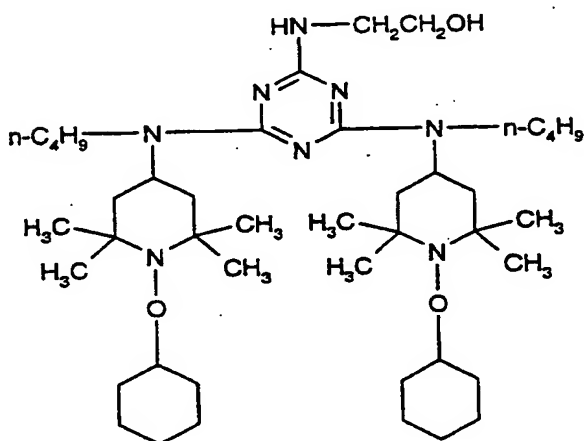
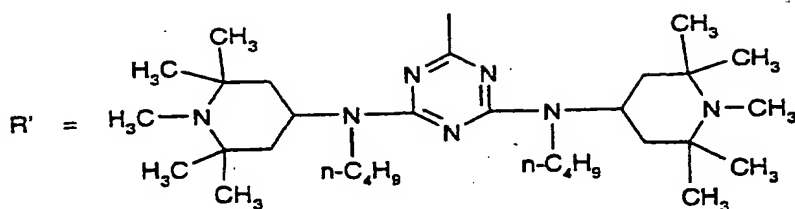
(H4) Tinuvin®622



(H5) Chimassorb®944



(H6) Chimassorb®119,



(H7)

in which

m is a number from the range from 2 to 200.

Component (e) of the novel composition, or the compounds of the sterically hindered amine type, are known and some are commercially available.

Tinuvin®123, Tinuvin®144, Tinuvin®292, Tinuvin®622, Chimassorb®944 und Chimassorb®119 are protected trade names of Ciba Spezialitätenchemie AG.

As component (e) of the novel composition particular preference is given to the addition of those sterically hindered amines whose molecular weight or average molecular weight  $M_n$  is in the range from 500 to 10 000, in particular in the range from 1000 to 10 000. Of these, particular emphasis should again be placed on those sterically hindered amines whose molecular weight or average molecular weight  $M_n$  is in the range from 1500 to 10 000, for example in the range from 2000 to 7500.

Particular emphasis should be given to those novel compositions which as component (e) comprise two or more compounds of the sterically hindered amine type.

The mixture of components (b), (c), (d) und (e) is suitable for stabilizing organic materials against oxidative, thermal or light-induced degradation. Examples of such materials are:

1. Polymers of monoolefins and diolefins, for example polypropylene, polyisobutylene, polybut-1-ene, poly-4-methylpent-1-ene, polyisoprene or polybutadiene, as well as polymers of cycloolefins, for instance of cyclopentene or norbornene, polyethylene (which optionally can be crosslinked), for example high density polyethylene (HDPE), high density and high molecular weight polyethylene (HDPE-HMW), high density and ultrahigh molecular weight polyethylene (HDPE-UHMW), medium density polyethylene (MDPE), low density polyethylene (LDPE), linear low density polyethylene (LLDPE), (VLDPE) and (ULDPE).

Polyolefins, i.e. the polymers of monoolefins exemplified in the preceding paragraph, preferably polyethylene and polypropylene, can be prepared by different, and especially by the following, methods:

- a) radical polymerisation (normally under high pressure and at elevated temperature).
- b) catalytic polymerisation using a catalyst that normally contains one or more than one metal of groups IVb, Vb, VIb or VIII of the Periodic Table. These metals usually have one or more than one ligand, typically oxides, halides, alcoholates, esters, ethers, amines, alkyls, alkenyls and/or aryls that may be either  $\pi$ - or  $\sigma$ -coordinated. These metal complexes may be in the free form or fixed on substrates, typically on activated magnesium chloride, titanium(III) chloride, alumina or silicon oxide. These catalysts may be soluble or insoluble in the polymerisation medium. The catalysts can be used by themselves in the polymerisation or further activators may be used, typically metal alkyls, metal hydrides, metal alkyl halides, metal alkyl oxides or metal alkyloxanes, said metals being elements of groups Ia, IIa and/or IIIa of the Periodic Table. The activators may be modified conveniently with further ester, ether, amine or silyl ether groups. These catalyst systems are usually termed Phillips, Standard Oil Indiana, Ziegler (-Natta), TNZ (DuPont), metallocene or single site catalysts (SSC).

2. Mixtures of the polymers mentioned under 1), for example mixtures of polypropylene with polyisobutylene, polypropylene with polyethylene (for example PP/HDPE, PP/LDPE) and mixtures of different types of polyethylene (for example LDPE/HDPE).
3. Copolymers of monoolefins and diolefins with each other or with other vinyl monomers, for example ethylene/propylene copolymers, linear low density polyethylene (LLDPE) and mixtures thereof with low density polyethylene (LDPE), propylene/but-1-ene copolymers, propylene/isobutylene copolymers, ethylene/but-1-ene copolymers, ethylene/hexene copolymers, ethylene/methylpentene copolymers, ethylene/heptene copolymers, ethylene/octene copolymers, propylene/butadiene copolymers, isobutylene/isoprene copolymers, ethylene/alkyl acrylate copolymers, ethylene/alkyl methacrylate copolymers, ethylene/vinyl acetate copolymers and their copolymers with carbon monoxide or ethylene/acrylic acid copolymers and their salts (ionomers) as well as terpolymers of ethylene with propylene and a diene such as hexadiene, dicyclopentadiene or ethylidene-norbornene; and mixtures of such copolymers with one another and with polymers mentioned in 1) above, for example polypropylene/ethylene-propylene copolymers, LDPE/ethylene-vinyl acetate copolymers (EVA), LDPE/ethylene-acrylic acid copolymers (EAA), LLDPE/EVA, LLDPE/EAA and alternating or random polyalkylene/carbon monoxide copolymers and mixtures thereof with other polymers, for example polyamides.
4. Hydrocarbon resins (for example  $C_5$ - $C_9$ ) including hydrogenated modifications thereof (e.g. tackifiers) and mixtures of polyalkylenes and starch.
5. Polystyrene, poly(p-methylstyrene), poly( $\alpha$ -methylstyrene).
6. Copolymers of styrene or  $\alpha$ -methylstyrene with dienes or acrylic derivatives, for example styrene/butadiene, styrene/acrylonitrile, styrene/alkyl methacrylate, styrene/butadiene/alkyl acrylate, styrene/butadiene/alkyl methacrylate, styrene/maleic anhydride, styrene/acrylonitrile/methyl acrylate; mixtures of high impact strength of styrene copolymers and another polymer, for example a polyacrylate, a diene polymer or an ethylene/propylene/diene terpolymer; and block copolymers of styrene such as styrene/butadiene/styrene, styrene/isoprene/styrene, styrene/ethylene/butylene/styrene or styrene/ethylene/propylene/styrene.

7. Graft copolymers of styrene or  $\alpha$ -methylstyrene, for example styrene on polybutadiene, styrene on polybutadiene-styrene or polybutadiene-acrylonitrile copolymers; styrene and acrylonitrile (or methacrylonitrile) on polybutadiene; styrene, acrylonitrile and methyl methacrylate on polybutadiene; styrene and maleic anhydride on polybutadiene; styrene, acrylonitrile and maleic anhydride or maleimide on polybutadiene; styrene and maleimide on polybutadiene; styrene and alkyl acrylates or methacrylates on polybutadiene; styrene and acrylonitrile on ethylene/propylene/diene terpolymers; styrene and acrylonitrile on polyalkyl acrylates or polyalkyl methacrylates, styrene and acrylonitrile on acrylate/butadiene copolymers, as well as mixtures thereof with the copolymers listed under 6), for example the copolymer mixtures known as ABS, MBS, ASA or AES polymers.
8. Halogen-containing polymers such as polychloroprene, chlorinated rubbers, chlorinated and brominated copolymer of isobutylene-isoprene (halobutyl rubber), chlorinated or sulfochlorinated polyethylene, copolymers of ethylene and chlorinated ethylene, epichlorohydrin homo- and copolymers, especially polymers of halogen-containing vinyl compounds, for example polyvinyl chloride, polyvinylidene chloride, polyvinyl fluoride, polyvinylidene fluoride, as well as copolymers thereof such as vinyl chloride/vinylidene chloride, vinyl chloride/vinyl acetate or vinylidene chloride/vinyl acetate copolymers.
9. Polymers derived from  $\alpha,\beta$ -unsaturated acids and derivatives thereof such as polyacrylates and polymethacrylates; polymethyl methacrylates, polyacrylamides and polyacrylonitriles, impact-modified with butyl acrylate.
10. Copolymers of the monomers mentioned under 9) with each other or with other unsaturated monomers, for example acrylonitrile/ butadiene copolymers, acrylonitrile/alkyl acrylate copolymers, acrylonitrile/alkoxyalkyl acrylate or acrylonitrile/vinyl halide copolymers or acrylonitrile/ alkyl methacrylate/butadiene terpolymers.
11. Polymers derived from unsaturated alcohols and amines or the acyl derivatives or acetals thereof, for example polyvinyl alcohol, polyvinyl acetate, polyvinyl stearate, polyvinyl benzoate, polyvinyl maleate, polyvinyl butyral, polyallyl phthalate or polyallyl melamine; as well as their copolymers with olefins mentioned in 1) above.



12. Homopolymers and copolymers of cyclic ethers such as polyalkylene glycols, polyethylene oxide, polypropylene oxide or copolymers thereof with bisglycidyl ethers.
13. Polyacetals such as polyoxymethylene and those polyoxymethylenes which contain ethylene oxide as a comonomer; polyacetals modified with thermoplastic polyurethanes, acrylates or MBS.
14. Polyphenylene oxides and sulfides, and mixtures of polyphenylene oxides with styrene polymers or polyamides.
15. Polyurethanes derived from hydroxyl-terminated polyethers, polyesters or polybutadienes on the one hand and aliphatic or aromatic polyisocyanates on the other, as well as precursors thereof.
16. Polyamides and copolyamides derived from diamines and dicarboxylic acids and/or from aminocarboxylic acids or the corresponding lactams, for example polyamide 4, polyamide 6, polyamide 6/6, 6/10, 6/9, 6/12, 4/6, 12/12, polyamide 11, polyamide 12, aromatic polyamides starting from m-xylene diamine and adipic acid; polyamides prepared from hexamethylenediamine and isophthalic or/and terephthalic acid and with or without an elastomer as modifier, for example poly-2,4,4'-trimethylhexamethylene terephthalamide or poly-m-phenylene isophthalamide; and also block copolymers of the aforementioned polyamides with polyolefins, olefin copolymers, ionomers or chemically bonded or grafted elastomers; or with polyethers, e.g. with polyethylene glycol, polypropylene glycol or polytetramethylene glycol; as well as polyamides or copolyamides modified with EPDM or ABS; and polyamides condensed during processing (RIM polyamide systems).
17. Polyureas, polyimides, polyamide-imides, polyetherimids, polyesterimids, polyhydantoins and polybenzimidazoles.
18. Polyesters derived from dicarboxylic acids and diols and/or from hydroxycarboxylic acids or the corresponding lactones, for example polyethylene terephthalate, polybutylene terephthalate, poly-1,4-dimethylolcyclohexane terephthalate and polyhydroxybenzoates, as well as block copolyether esters derived from hydroxyl-terminated polyethers; and also polyesters modified with polycarbonates or MBS.

19. Polycarbonates and polyester carbonates.
20. Polysulfones, polyether sulfones and polyether ketones.
21. Crosslinked polymers derived from aldehydes on the one hand and phenols, ureas and melamines on the other hand, such as phenol/formaldehyde resins, urea/formaldehyde resins and melamine/formaldehyde resins.
22. Drying and non-drying alkyd resins.
23. Unsaturated polyester resins derived from copolyesters of saturated and unsaturated dicarboxylic acids with polyhydric alcohols and vinyl compounds as crosslinking agents, and also halogen-containing modifications thereof of low flammability.
24. Crosslinkable acrylic resins derived from substituted acrylates, for example epoxy acrylates, urethane acrylates or polyester acrylates.
25. Alkyd resins, polyester resins and acrylate resins crosslinked with melamine resins, urea resins, isocyanates, isocyanurates, polyisocyanates or epoxy resins.
26. Crosslinked epoxy resins derived from aliphatic, cycloaliphatic, heterocyclic or aromatic glycidyl compounds, e.g. products of diglycidyl ethers of bisphenol A and bisphenol F, which are crosslinked with customary hardeners such as anhydrides or amines, with or without accelerators.
27. Natural polymers such as cellulose, rubber, gelatin and chemically modified homologous derivatives thereof, for example cellulose acetates, cellulose propionates and cellulose butyrates, or the cellulose ethers such as methyl cellulose; as well as rosins and their derivatives.
28. Blends of the aforementioned polymers (polyblends), for example PP/EPDM, Polyamide/EPDM or ABS, PVC/EVA, PVC/ABS, PVC/MBS, PC/ABS, PBTP/ABS, PC/ASA, PC/PBT, PVC/CPE, PVC/acrylates, POM/thermoplastic PUR, PC/thermoplastic PUR,

POM/acrylate, POM/MBS, PPO/HIPS, PPO/PA 6.6 and copolymers, PA/HDPE, PA/PP, PA/PPO, PBT/PC/ABS or PBT/PET/PC.

29. Naturally occurring and synthetic organic materials which are pure monomeric compounds or mixtures of such compounds, for example mineral oils, animal and vegetable fats, oil and waxes, or oils, fats and waxes based on synthetic esters (e.g. phthalates, adipates, phosphates or trimellitates) and also mixtures of synthetic esters with mineral oils in any weight ratios, typically those used as spinning compositions, as well as aqueous emulsions of such materials.

30. Aqueous emulsions of natural or synthetic rubber, e.g. natural latex or latices of carboxylated styrene/butadiene copolymers.

The mixture of components (b), (c), (d) and (e) is likewise used for polyurethane production, especially for preparing flexible polyurethane foams. In this context the novel compositions and the products produced therefrom are effectively protected against degradation. In particular, scorching during foam production is avoided.

The polyurethanes are obtained, for example, by reacting polyethers, polyesters and polybutadienes which contain terminal hydroxyl groups with aliphatic or aromatic polyisocyanates.

Polyethers having terminal hydroxyl groups are known and are prepared, for example, by polymerizing epoxides such as ethylene oxide, propylene oxide, butylene oxide, tetrahydrofuran, styrene oxide or epichlorohydrin with themselves, for example in the presence of  $\text{BF}_3$ , or by addition reaction of these epoxides, alone or as a mixture or in succession, with starting components containing reactive hydrogen atoms, such as water, alcohols, ammonia or amines, for example ethylene glycol, propylene 1,3- and 1,2-glycol, trimethylolpropane, 4,4'-dihydroxydiphenylpropane, aniline, ethanolamine or ethylenediamine. Sucrose polyethers are also suitable in accordance with the invention. In many cases preference is given to those polyethers which predominantly (up to 90% by weight, based on all the OH groups present in the polyether) contain primary OH groups. Furthermore, polyethers modified by vinyl polymers, as are formed, for example, by polymerizing styrene and acrylonitrile in the presence of polyethers, are suitable, as are polybutadienes containing OH groups.

These compounds generally have molecular weights of 400-10000 and are polyhydroxy compounds, especially compounds containing from two to eight hydroxyl groups, especially those of molecular weight from 800 to 10 000, preferably from 1000 to 6000, for example polyethers containing at least 2, generally 2 to 8, but preferably 2 to 4, hydroxyl groups, as are known per se for the preparation of homogeneous polyurethanes and cellular polyurethanes.

It is of course possible to employ mixtures of the above compounds containing at least two isocyanate-reactive hydrogen atoms, in particular with a molecular weight of 400 - 10 000.

Suitable polyisocyanates are aliphatic, cycloaliphatic, araliphatic, aromatic and heterocyclic polyisocyanates, for example ethylene diisocyanate, 1,4-tetramethylene diisocyanate, 1,6-hexamethylene diisocyanate, 1,12-dodecane diisocyanate, cyclobutane 1,3-diisocyanate, cyclohexane 1,3- and -1,4-diisocyanate and also any desired mixtures of these isomers, 1-isocyanato-3,3,5-trimethyl-5-isocyanatomethylcyclohexane, 2,4- and 2,6-hexahydrotolylene diisocyanate and also any desired mixtures of these isomers, hexahydro-1,3- and/or -1,4-phenylene diisocyanate, perhydro-2,4'- and/or -4,4'-diphenylmethanediisocyanate, 1,3- and 1,4-phenylene diisocyanate, 2,4- and 2,6-tolylene diisocyanate, and also any desired mixtures of these isomers, diphenylmethane 2,4'- and/or -4,4'-diisocyanate, naphthylene 1,5-diisocyanate, triphenylmethane 4,4',4"-triisocyanate, polyphenyl-polymethylene polyisocyanates as are obtained by aniline-formaldehyde condensation followed by phosgenization, m- and p-isocyanatophenylsulfonyl isocyanates, perchlorinated aryl polyisocyanates, polyisocyanates containing carbodiimide groups, polyisocyanates containing allophanate groups, polyisocyanates containing isocyanurate groups, polyisocyanates containing urethane groups, polyisocyanates containing acylated urea groups, polyisocyanates containing biuret groups, polyisocyanates containing ester groups, reaction products of the abovementioned isocyanates with acetals, and polyisocyanates containing polymeric fatty acid radicals.

It is also possible to employ the isocyanate group-containing distillation residues, as they are or dissolved in one or more of the abovementioned polyisocyanates, which are obtained in the course of the industrial preparation of isocyanates. It is additionally possible to use any desired mixtures of the abovementioned polyisocyanates.

Particular preference is given in general to the polyisocyanates which are readily obtainable industrially, for example 2,4- and 2,6-tolylene diisocyanate and any desired mixtures of these isomers ("TDI"), polyphenyl-polymethylene-polyisocyanates as prepared by aniline-formaldehyde condensation followed by phosgenization ("crude MDI"), and polyisocyanates containing carbodiimide, urethane, allophanate, isocyanurate, urea or biuret groups ("modified polyisocyanates").

The organic materials to be protected are preferably natural, semisynthetic or preferably synthetic polymers. Particular preference is given to thermoplastic polymers, especially polyolefins, in particular polyethylene and polypropylene or copolymers thereof with mono- and diolefins.

Particular emphasis is to be placed on the action of the novel compounds against thermal and oxidative degradation, especially under thermal stress as occurs during the processing of thermoplastics. The novel components (b), (c), (d) and (e) are therefore outstandingly suitable for use as processing (in-process) stabilizers.

The mixture of components (b), (c), (d) and (e) is also suitable for stabilizing polyolefins which are in long-term contact with extracting media.

Component (b) is preferably added to the organic material to be stabilized in an amount of from 0.0005 to 5%, in particular from 0.001 to 2%, for example from 0.01 to 2%, based on the weight of the organic material to be stabilized.

Components (c), (d) and (e) are judiciously added to the organic material to be stabilized in an amount of from 0.01 to 10%, for example from 0.01 to 5%, preferably from 0.025 to 3% and, in particular, from 0.025 to 1%, based on the weight of the organic material to be stabilized.

In addition to components (a), (b), (c), (d) and (e) the novel compositions may comprise further costabilizers (additives) the following, for example:

#### 1. Antioxidants

1.1. Alkylated monophenols, for example 2,6-di-tert-butyl-4-methylphenol, 2-tert-butyl-4,6-dimethylphenol, 2,6-di-tert-butyl-4-ethylphenol, 2,6-di-tert-butyl-4-n-butylphenol, 2,6-di-tert-butyl-4-isobutylphenol, 2,6-dicyclopentyl-4-methylphenol, 2-( $\alpha$ -methylcyclohexyl)-4,6-dimethylphenol, 2,6-dioctadecyl-4-methylphenol, 2,4,6-tricyclohexylphenol, 2,6-di-tert-butyl-4-methoxymethylphenol, nonylphenols which are linear or branched in the side chains, for example, 2,6-di-nonyl-4-methylphenol, 2,4-dimethyl-6-(1'-methylundec-1'-yl)phenol, 2,4-dimethyl-6-(1'-methylheptadec-1'-yl)phenol, 2,4-dimethyl-6-(1'-methyltridec-1'-yl)phenol and mixtures thereof.

1.2. Alkylthiomethylphenols, for example 2,4-dioctylthiomethyl-6-tert-butylphenol, 2,4-dioctylthiomethyl-6-methylphenol, 2,4-dioctylthiomethyl-6-ethylphenol, 2,6-di-dodecylthiomethyl-4-nonylphenol.

1.3. Hydroquinones and alkylated hydroquinones, for example 2,6-di-tert-butyl-4-methoxyphenol, 2,5-di-tert-butylhydroquinone, 2,5-di-tert-amylhydroquinone, 2,6-diphenyl-4-octadecyloxyphenol, 2,6-di-tert-butylhydroquinone, 2,5-di-tert-butyl-4-hydroxyanisole, 3,5-di-tert-butyl-4-hydroxyanisole, 3,5-di-tert-butyl-4-hydroxyphenyl stearate, bis-(3,5-di-tert-butyl-4-hydroxyphenyl) adipate.

1.4. Tocopherols, for example  $\alpha$ -tocopherol,  $\beta$ -tocopherol,  $\gamma$ -tocopherol,  $\delta$ -tocopherol and mixtures thereof (Vitamin E).

1.5. Hydroxylated thiodiphenyl ethers, for example 2,2'-thiobis(6-tert-butyl-4-methylphenol), 2,2'-thiobis(4-octylphenol), 4,4'-thiobis(6-tert-butyl-3-methylphenol), 4,4'-thiobis(6-tert-butyl-2-methylphenol), 4,4'-thiobis-(3,6-di-sec-amylphenol), 4,4'-bis(2,6-dimethyl-4-hydroxyphenyl)disulfide.

1.6. Alkylidenebisphenols, for example 2,2'-methylenebis(6-tert-butyl-4-methylphenol), 2,2'-methylenebis(6-tert-butyl-4-ethylphenol), 2,2'-methylenebis[4-methyl-6-( $\alpha$ -methylcyclohexyl)phenol], 2,2'-methylenebis(4-methyl-6-cyclohexylphenol), 2,2'-methylenebis(6-nonyl-4-methylphenol), 2,2'-methylenebis(4,6-di-tert-butylphenol), 2,2'-ethylidenebis(4,6-di-tert-butylphenol), 2,2'-ethylidenebis(6-tert-butyl-4-isobutylphenol), 2,2'-methylenebis[6-( $\alpha$ -methylbenzyl)-4-nonylphenol], 2,2'-methylenebis[6-( $\alpha,\alpha$ -dimethylbenzyl)-4-nonylphenol], 4,4'-methylenebis(2,6-di-tert-butylphenol), 4,4'-methylenebis(6-tert-butyl-2-methylphenol), 1,1-bis(5-

tert-butyl-4-hydroxy-2-methylphenyl)butane, 2,6-bis(3-tert-butyl-5-methyl-2-hydroxybenzyl)-4-methylphenol, 1,1,3-tris(5-tert-butyl-4-hydroxy-2-methylphenyl)butane, 1,1-bis(5-tert-butyl-4-hydroxy-2-methylphenyl)-3-n-dodecylmercaptobutane, ethylene glycol bis[3,3-bis(3'-tert-butyl-4'-hydroxyphenyl)butyrate], bis(3-tert-butyl-4-hydroxy-5-methylphenyl)dicyclopentadiene, bis[2-(3'-tert-butyl-2'-hydroxy-5'-methylbenzyl)-6-tert-butyl-4-methylphenyl]terephthalate, 1,1-bis-(3,5-dimethyl-2-hydroxyphenyl)butane, 2,2-bis-(3,5-di-tert-butyl-4-hydroxyphenyl)propane, 2,2-bis-(5-tert-butyl-4-hydroxy-2-methylphenyl)-4-n-dodecylmercaptobutane, 1,1,5,5-tetra-(5-tert-butyl-4-hydroxy-2-methylphenyl)pentane.

1.7. O-, N- and S-benzyl compounds, for example 3,5,3',5'-tetra-tert-butyl-4,4'-dihydroxydibenzyl ether, octadecyl-4-hydroxy-3,5-dimethylbenzylmercaptoacetate, tridecyl-4-hydroxy-3,5-di-tert-butylbenzylmercaptoacetate, tris(3,5-di-tert-butyl-4-hydroxybenzyl)amine, bis(4-tert-butyl-3-hydroxy-2,6-dimethylbenzyl)dithioterephthalate, bis(3,5-di-tert-butyl-4-hydroxybenzyl)sulfide, isooctyl-3,5-di-tert-butyl-4-hydroxybenzylmercaptoacetate.

1.8. Hydroxybenzylated malonates, for example dioctadecyl-2,2-bis-(3,5-di-tert-butyl-2-hydroxybenzyl)-malonate, di-octadecyl-2-(3-tert-butyl-4-hydroxy-5-methylbenzyl)-malonate, di-dodecylmercaptoethyl-2,2-bis-(3,5-di-tert-butyl-4-hydroxybenzyl)malonate, bis[4-(1,1,3,3-tetramethylbutyl)phenyl]-2,2-bis(3,5-di-tert-butyl-4-hydroxybenzyl)malonate.

1.9. Aromatic hydroxybenzyl compounds, for example 1,3,5-tris-(3,5-di-tert-butyl-4-hydroxybenzyl)-2,4,6-trimethylbenzene, 1,4-bis(3,5-di-tert-butyl-4-hydroxybenzyl)-2,3,5,6-tetramethylbenzene, 2,4,6-tris(3,5-di-tert-butyl-4-hydroxybenzyl)phenol.

1.10. Triazine Compounds, for example 2,4-bis(octylmercapto)-6-(3,5-di-tert-butyl-4-hydroxyanilino)-1,3,5-triazine, 2-octylmercapto-4,6-bis(3,5-di-tert-butyl-4-hydroxyanilino)-1,3,5-triazine, 2-octylmercapto-4,6-bis(3,5-di-tert-butyl-4-hydroxyphenoxy)-1,3,5-triazine, 2,4,6-tris-(3,5-di-tert-butyl-4-hydroxyphenoxy)-1,2,3-triazine, 1,3,5-tris-(3,5-di-tert-butyl-4-hydroxybenzyl)isocyanurate, 1,3,5-tris(4-tert-butyl-3-hydroxy-2,6-dimethylbenzyl)isocyanurate, 2,4,6-tris-(3,5-di-tert-butyl-4-hydroxyphenylethyl)-1,3,5-triazine, 1,3,5-tris(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)-hexahydro-1,3,5-triazine, 1,3,5-tris(3,5-dicyclohexyl-4-hydroxybenzyl)isocyanurate.

1.11. Benzylphosphonates, for example dimethyl-2,5-di-tert-butyl-4-hydroxybenzylphosphonate, diethyl-3,5-di-tert-butyl-4-hydroxybenzylphosphonate, dioctadecyl-3,5-di-tert-butyl-4-hydroxybenzylphosphonate, dioctadecyl-5-tert-butyl-4-hydroxy-3-methylbenzylphosphonate, the calcium salt of the monoethyl ester of 3,5-di-tert-butyl-4-hydroxybenzylphosphonic acid.

1.12. Acylaminophenols, for example 4-hydroxylauranilide, 4-hydroxystearanilide, octyl N-(3,5-di-tert-butyl-4-hydroxyphenyl)carbamate.

1.13. Esters of  $\beta$ -(3,5-di-tert-butyl-4-hydroxyphenyl)propionic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, n-octanol, i-octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl) isocyanurate, N,N'-bis(hydroxyethyl)oxamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane.

1.14. Esters of  $\beta$ -(5-tert-butyl-4-hydroxy-3-methylphenyl)propionic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, n-octanol, i-octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl) isocyanurate, N,N'-bis(hydroxyethyl)oxamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane.

1.15. Esters of  $\beta$ -(3,5-dicyclohexyl-4-hydroxyphenyl)propionic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl)isocyanurate, N,N'-bis(hydroxyethyl)oxamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane.

1.16. Esters of 3,5-di-tert-butyl-4-hydroxyphenyl acetic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl)isocyanurate, N,N'-bis(hydroxyethyl)ox-



amide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane.

1.17. Amides of  $\beta$ -(3,5-di-tert-butyl-4-hydroxyphenyl)propionic acid e.g. N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)hexamethylenediamide, N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)trimethylenediamide, N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)hydrazide, N,N'-bis[2-(3-[3,5-di-tert-butyl-4-hydroxyphenyl]propionyloxy)ethyl]oxamide (Naugard®XL-1 supplied by Uniroyal).

1.18. Ascorbic acid (vitamin C)

1.19. Aminic antioxidants, for example N,N'-di-isopropyl-p-phenylenediamine, N,N'-di-sec-butyl-p-phenylenediamine, N,N'-bis(1,4-dimethylpentyl)-p-phenylenediamine, N,N'-bis(1-ethyl-3-methylpentyl)-p-phenylenediamine, N,N'-bis(1-methylheptyl)-p-phenylenediamine, N,N'-dicyclohexyl-p-phenylenediamine, N,N'-diphenyl-p-phenylenediamine, N,N'-bis(2-naphthyl)-p-phenylenediamine, N-isopropyl-N'-phenyl-p-phenylenediamine, N-(1,3-dimethylbutyl)-N'-phenyl-p-phenylenediamine, N-(1-methylheptyl)-N'-phenyl-p-phenylenediamine, N-cyclohexyl-N'-phenyl-p-phenylenediamine, 4-(p-toluenesulfamoyl)diphenylamine, N,N'-dimethyl-N,N'-di-sec-butyl-p-phenylenediamine, diphenylamine, N-allyldiphenylamine, 4-isopropoxydiphenylamine, N-phenyl-1-naphthylamine, N-(4-tert-octylphenyl)-1-naphthylamine, N-phenyl-2-naphthylamine, octylated diphenylamine, for example p,p'-di-tert-octyldiphenylamine, 4-n-butylaminophenol, 4-butyrylaminophenol, 4-nonanoylaminophenol, 4-dodecanoylaminophenol, 4-octadecanoylaminophenol, bis(4-methoxyphenyl)amine, 2,6-di-tert-butyl-4-dimethylaminomethylphenol, 2,4'-diaminodiphenylmethane, 4,4'-diaminodiphenylmethane, N,N,N',N'-tetramethyl-4,4'-diaminodiphenylmethane, 1,2-bis[(2-methylphenyl)amino]ethane, 1,2-bis(phenylamino)propane, (o-tolyl)biguanide, bis[4-(1',3'-dimethylbutyl)phenyl]amine, tert-octylated N-phenyl-1-naphthylamine, a mixture of mono- and dialkylated tert-butyl/tert-octyldiphenylamines, a mixture of mono- and dialkylated nonyldiphenylamines, a mixture of mono- and dialkylated dodecyldiphenylamines, a mixture of mono- and dialkylated isopropyl/isohexyldiphenylamines, a mixture of mono- und dialkylated tert-butyl/diphenylamines, 2,3-dihydro-3,3-dimethyl-4H-1,4-benzothiazine, phenothiazine, a mixture of mono- und dialkylated tert-butyl/tert-octylphenothiazines, a mixture of mono- und dialkylated tert-octyl-phenothiazines, N-allylphenothiazin, N,N,N',N'-tetraphenyl-1,4-diaminobut-2-ene, N,N-bis-

(2,2,6,6-tetramethyl-piperid-4-yl-hexamethylenediamine, bis(2,2,6,6-tetramethylpiperid-4-yl)-sebacate, 2,2,6,6-tetramethylpiperidin-4-one, 2,2,6,6-tetramethylpiperidin-4-ol.

## 2. UV absorbers and light stabilisers

2.1. 2-(2'-Hydroxyphenyl)benzotriazoles, for example 2-(2'-hydroxy-5'-methylphenyl)-benzotriazole, 2-(3',5'-di-tert-butyl-2'-hydroxyphenyl)benzotriazole, 2-(5'-tert-butyl-2'-hydroxyphenyl)benzotriazole, 2-(2'-hydroxy-5'-(1,1,3,3-tetramethylbutyl)phenyl)benzotriazole, 2-(3',5'-di-tert-butyl-2'-hydroxyphenyl)-5-chloro-benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-methylphenyl)-5-chloro-benzotriazole, 2-(3'-sec-butyl-5'-tert-butyl-2'-hydroxyphenyl)benzotriazole, 2-(2'-hydroxy-4'-octyloxyphenyl)benzotriazole, 2-(3',5'-di-tert-amyl-2'-hydroxyphenyl)benzotriazole, 2-(3',5'-bis-( $\alpha,\alpha$ -dimethylbenzyl)-2'-hydroxyphenyl)benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-octyloxycarbonylethyl)phenyl)-5-chloro-benzotriazole, 2-(3'-tert-butyl-5'-[2-(2-ethylhexyloxy)-carbonylethyl]-2'-hydroxyphenyl)-5-chloro-benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-methoxycarbonylethyl)phenyl)-5-chloro-benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-methoxycarbonylethyl)phenyl)benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-octyloxycarbonylethyl)phenyl)benzotriazole, 2-(3'-tert-butyl-5'-[2-(2-ethylhexyloxy)carbonylethyl]-2'-hydroxyphenyl)benzotriazole, 2-(3'-dodecyl-2'-hydroxy-5'-methylphenyl)benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-isooctyloxycarbonylethyl)phenyl)benzotriazole, 2,2'-methylene-bis[4-(1,1,3,3-tetramethylbutyl)-6-benzotriazole-2-ylphenol]; the transesterification product of 2-[3'-tert-butyl-5'-(2-methoxycarbonylethyl)-2'-hydroxyphenyl]-2H-benzotriazole with polyethylene glycol 300:  $\left[ R-CH_2CH_2-COO-CH_2CH_2 \right]_2$  where R = 3'-tert-butyl-4'-hydroxy-5'-2H-benzotriazole-2-ylphenyl, 2-[2'-hydroxy-3'-( $\alpha,\alpha$ -dimethylbenzyl)-5'-(1,1,3,3-tetramethylbutyl)-phenyl]benzotriazole; 2-[2'-hydroxy-3'-(1,1,3,3-tetramethylbutyl)-5'-( $\alpha,\alpha$ -dimethylbenzyl)-phenyl]benzotriazole.

2.2. 2-Hydroxybenzophenones, for example the 4-hydroxy, 4-methoxy, 4-octyloxy, 4-decyloxy, 4-dodecyloxy, 4-benzyloxy, 4,2',4'-trihydroxy and 2'-hydroxy-4,4'-dimethoxy derivatives.

2.3. Esters of substituted and unsubstituted benzoic acids, as for example 4-tertbutyl-phenyl salicylate, phenyl salicylate, octylphenyl salicylate, dibenzoyl resorcinol, bis(4-tert-butylbenzoyl) resorcinol, benzoyl resorcinol, 2,4-di-tert-butylphenyl 3,5-di-tert-butyl-4-hydroxybenzo-

ate, hexadecyl 3,5-di-tert-butyl-4-hydroxybenzoate, octadecyl 3,5-di-tert-butyl-4-hydroxybenzoate, 2-methyl-4,6-di-tert-butylphenyl 3,5-di-tert-butyl-4-hydroxybenzoate.

2.4. Acrylates, for example ethyl  $\alpha$ -cyano- $\beta,\beta$ -diphenylacrylate, isooctyl  $\alpha$ -cyano- $\beta,\beta$ -diphenylacrylate, methyl  $\alpha$ -carbomethoxycinnamate, methyl  $\alpha$ -cyano- $\beta$ -methyl-p-methoxy-cinnamate, butyl  $\alpha$ -cyano- $\beta$ -methyl-p-methoxy-cinnamate, methyl  $\alpha$ -carbomethoxy-p-methoxycinnamate and N-( $\beta$ -carbomethoxy- $\beta$ -cyanovinyl)-2-methylindoline.

2.5. Nickel compounds, for example nickel complexes of 2,2'-thio-bis-[4-(1,1,3,3-tetramethylbutyl)phenol], such as the 1:1 or 1:2 complex, with or without additional ligands such as n-butylamine, triethanolamine or N-cyclohexyldiethanolamine, nickel dibutyldithiocarbamate, nickel salts of the monoalkyl esters, e.g. the methyl or ethyl ester, of 4-hydroxy-3,5-di-tert-butylbenzylphosphonic acid, nickel complexes of ketoximes, e.g. of 2-hydroxy-4-methylphenyl undecylketoxime, nickel complexes of 1-phenyl-4-lauroyl-5-hydroxypyrazole, with or without additional ligands.

2.6. Sterically hindered amines, for example bis(2,2,6,6-tetramethyl-4-piperidyl)sebacate, bis(2,2,6,6-tetramethyl-4-piperidyl)succinate, bis(1,2,2,6,6-pentamethyl-4-piperidyl)sebacate, bis(1-octyloxy-2,2,6,6-tetramethyl-4-piperidyl)sebacate, bis(1,2,2,6,6-pentamethyl-4-piperidyl) n-butyl-3,5-di-tert-butyl-4-hydroxybenzylmalonate, the condensate of 1-(2-hydroxyethyl)-2,2,6,6-tetramethyl-4-hydroxypiperidine and succinic acid, linear or cyclic condensates of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine and 4-tert-octylamino-2,6-dichloro-1,3,5-triazine, tris(2,2,6,6-tetramethyl-4-piperidyl)nitrilotriacetate, tetrakis(2,2,6,6-tetramethyl-4-piperidyl)-1,2,3,4-butane-tetracarboxylate, 1,1'-(1,2-ethanediyl)-bis(3,3,5,5-tetramethylpiperazinone), 4-benzoyl-2,2,6,6-tetramethylpiperidine, 4-stearyloxy-2,2,6,6-tetramethylpiperidine, bis(1,2,2,6,6-pentamethylpiperidyl)-2-n-butyl-2-(2-hydroxy-3,5-di-tert-butylbenzyl)malonate, 3-n-octyl-7,7,9,9-tetramethyl-1,3,8-triazaspiro[4.5]decan-2,4-dione, bis(1-octyloxy-2,2,6,6-tetramethylpiperidyl)sebacate, bis(1-octyloxy-2,2,6,6-tetramethylpiperidyl)succinate, linear or cyclic condensates of N,N'-bis-(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine and 4-morpholino-2,6-dichloro-1,3,5-triazine, the condensate of 2-chloro-4,6-bis(4-n-butylamino-2,2,6,6-tetramethylpiperidyl)-1,3,5-triazine and 1,2-bis(3-aminopropylamino)ethane, the condensate of 2-chloro-4,6-di-(4-n-butylamino-1,2,2,6,6-pentamethylpiperidyl)-1,3,5-triazine and 1,2-bis-(3-aminopropylamino)ethane, 8-acetyl-3-dodecyl-7,7,9,9-tetramethyl-1,3,8-triazaspiro[4.5]decane-2,4-dione, 3-dodecyl-1-(2,2,6,6-tetrame-

thyl-4-piperidyl)pyrrolidin-2,5-dione, 3-dodecyl-1-(1,2,2,6,6-pentamethyl-4-piperidyl)pyrrolidin-2,5-dione, a mixture of 4-hexadecyloxy- and 4-stearyloxy-2,2,6,6-tetramethylpiperidine, a condensation product of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine and 4-cyclohexylamino-2,6-dichloro-1,3,5-triazine, a condensation product of 1,2-bis(3-aminopropylamino)ethane and 2,4,6-trichloro-1,3,5-triazine as well as 4-butylamino-2,2,6,6-tetramethylpiperidine (CAS Reg. No. [136504-96-6]); N-(2,2,6,6-tetramethyl-4-piperidyl)-n-dodecylsuccinimid, N-(1,2,2,6,6-pentamethyl-4-piperidyl)-n-dodecylsuccinimid, 2-undecyl-7,7,9,9-tetramethyl-1-oxa-3,8-diaza-4-oxo-spiro[4,5]decane, a reaction product of 7,7,9,9-tetramethyl-2-cycloundecyl-1-oxa-3,8-diaza-4-oxospiro [4,5]decane und epichlorohydrin, 1,1-bis(1,2,2,6,6-pentamethyl-4-piperidyloxy-carbonyl)-2-(4-methoxyphenyl)ethene, N,N'-bis-formyl-N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine, diester of 4-methoxymethylene-malonic acid with 1,2,2,6,6-pentamethyl-4-hydroxypiperidine, poly[methylpropyl-3-oxy-4-(2,2,6,6-tetramethyl-4-piperidyl)]siloxane, reaction product of maleic acid anhydride- $\alpha$ -olefin-copolymer with 2,2,6,6-tetramethyl-4-aminopiperidine or 1,2,2,6,6-pentamethyl-4-aminopiperidine.

2.7. Oxamides, for example 4,4'-dioctyloxyoxanilide, 2,2'-diethoxyoxanilide, 2,2'-dioctyloxy-5,5'-di-tert-butoxanilide, 2,2'-didodecyloxy-5,5'-di-tert-butoxanilide, 2-ethoxy-2'-ethyloxanilide, N,N'-bis(3-dimethylaminopropyl)oxamide, 2-ethoxy-5-tert-butyl-2'-ethoxanilide and its mixture with 2-ethoxy-2'-ethyl-5,4'-di-tert-butoxanilide, mixtures of o- and p-methoxy-disubstituted oxanilides and mixtures of o- and p-ethoxy-disubstituted oxanilides.

2.8. 2-(2-Hydroxyphenyl)-1,3,5-triazines, for example 2,4,6-tris(2-hydroxy-4-octyloxyphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-octyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2,4-dihydroxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2,4-bis(2-hydroxy-4-propyloxyphenyl)-6-(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-octyloxyphenyl)-4,6-bis(4-methylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-dodecyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-tridecyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-[2-hydroxy-4-(2-hydroxy-3-butyloxy-propoxy)phenyl]-4,6-bis(2,4-dimethyl)-1,3,5-triazine, 2-[2-hydroxy-4-(2-hydroxy-3-octyloxy-propyloxy)phenyl]-4,6-bis(2,4-dimethyl)-1,3,5-triazine, 2-[4-(dodecyloxy/tridecyloxy-2-hydroxypropoxy)-2-hydroxy-phenyl]-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-[2-hydroxy-4-(2-hydroxy-3-dodecyloxy-propoxy)phenyl]-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-hexyloxy)phenyl-4,6-diphenyl-1,3,5-triazine, 2-(2-hydroxy-4-methoxyphenyl)-4,6-diphenyl-1,3,5-triazine, 2,4,6-tris[2-hydroxy-4-(3-

butoxy-2-hydroxy-propoxy)phenyl]-1,3,5-triazine, 2-(2-hydroxyphenyl)-4-(4-methoxyphenyl)-6-phenyl-1,3,5-triazine, 2-{2-hydroxy-4-[3-(2-ethylhexyl-1-oxy)-2-hydroxypropyloxy]phenyl}-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine.

3. Metal deactivators, for example N,N'-diphenyloxamide, N-salicylal-N'-salicyloyl hydrazine, N,N'-bis(salicyloyl) hydrazine, N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl) hydrazine, 3-salicyloylamino-1,2,4-triazole, bis(benzylidene)oxalyl dihydrazide, oxanilide, isophthaloyl dihydrazide, sebacoyl bisphenylhydrazide, N,N'-diacetyl adipoyl dihydrazide, N,N'-bis(salicyloyl)oxalyl dihydrazide, N,N'-bis(salicyloyl)thiopropionyl dihydrazide.

4. Phosphites and phosphonites, for example triphenyl phosphite, diphenyl alkyl phosphites, phenyl dialkyl phosphites, tris(nonylphenyl) phosphite, trilauryl phosphite, trioctadecyl phosphite, distearyl pentaerythritol diphosphite, tris(2,4-di-tert-butylphenyl) phosphite, diisodecyl pentaerythritol diphosphite, bis(2,4-di-tert-butylphenyl) pentaerythritol diphosphite, bis(2,6-di-tert-butyl-4-methylphenyl)-pentaerythritol diphosphite, diisodecyloxypentaerythritol diphosphite, bis(2,4-di-tert-butyl-6-methylphenyl)pentaerythritol diphosphite, bis(2,4,6-tris(tert-butylphenyl)pentaerythritol diphosphite, tristearyl sorbitol triphosphite, tetrakis(2,4-di-tert-butylphenyl) 4,4'-biphenylene diphosphonite, 6-isooctyloxy-2,4,8,10-tetra-tert-butyl-12H-dibenz[d,g]-1,3,2-dioxaphosphocin, 6-fluoro-2,4,8,10-tetra-tert-butyl-12-methyl-dibenz[d,g]-1,3,2-dioxaphosphocin, bis(2,4-di-tert-butyl-6-methylphenyl) methyl phosphite, bis(2,4-di-tert-butyl-6-methylphenyl) ethyl phosphite, 2,2',2''-nitriolo[triethyltris(3,3',5,5'-tetra-tert-butyl-1,1'-biphenyl-2,2'-diyl)phosphite], 2-ethylhexyl(3,3',5,5'-tetra-tert-butyl-1,1'-biphenyl-2,2'-diyl)phosphite.

5. Hydroxylamines, for example, N,N-dibenzylhydroxylamine, N,N-diethylhydroxylamine, N,N-dioctylhydroxylamine, N,N-dilaurylhydroxylamine, N,N-ditetradecylhydroxylamine, N,N-dihexadecylhydroxylamine, N,N-dioctadecylhydroxylamine, N-hexadecyl-N-octadecylhydroxylamine, N-heptadecyl-N-octadecylhydroxylamine, N,N-dialkylhydroxylamine derived from hydrogenated tallow amine.

6. Nitrones, for example, N-benzyl-alpha-phenyl-nitrone, N-ethyl-alpha-methyl-nitrone, N-octyl-alpha-heptyl-nitrone, N-lauryl-alpha-undecyl-nitrone, N-tetradecyl-alpha-tridcyl-nitrone, N-hexadecyl-alpha-pentadecyl-nitrone, N-octadecyl-alpha-heptadecyl-nitrone, N-hexadecyl-alpha-heptadecyl-nitrone, N-octadecyl-alpha-pentadecyl-nitrone, N-heptadecyl-alpha-hep-

tadecyl-nitrone, N-octadecyl-alpha-hexadecyl-nitrone, nitrone derived from N,N-dialkylhydroxylamine derived from hydrogenated tallow amine.

7. Thiosynergists, for example, dilauryl thiodipropionate or distearyl thiodipropionate.

8. Peroxide scavengers, for example esters of  $\beta$ -thiodipropionic acid, for example the lauryl, stearyl, myristyl or tridecyl esters, mercaptobenzimidazole or the zinc salt of 2-mercaptobenzimidazole, zinc dibutyldithiocarbamate, dioctadecyl disulfide, pentaerythritol tetrakis( $\beta$ -dodecylmercapto)propionate.

9. Polyamide stabilisers, for example, copper salts in combination with iodides and/or phosphorus compounds and salts of divalent manganese.

10. Basic co-stabilisers, for example, melamine, polyvinylpyrrolidone, dicyandiamide, triallyl cyanurate, urea derivatives, hydrazine derivatives, amines, polyamides, polyurethanes, alkali metal salts and alkaline earth metal salts of higher fatty acids for example calcium stearate, zinc stearate, magnesium behenate, magnesium stearate, sodium ricinoleate and potassium palmitate, antimony pyrocatecholate or zinc pyrocatecholate.

11. Nucleating agents, for example, inorganic substances such as talcum, metal oxides such as titanium dioxide or magnesium oxide, phosphates, carbonates or sulfates of, preferably, alkaline earth metals; organic compounds such as mono- or polycarboxylic acids and the salts thereof, e.g. 4-tert-butylbenzoic acid, adipic acid, diphenylacetic acid, sodium succinate or sodium benzoate; polymeric compounds such as ionic copolymers (ionomers).

12. Fillers and reinforcing agents, for example, calcium carbonate, silicates, glass fibres, glass bulbs, asbestos, talc, kaolin, mica, barium sulfate, metal oxides and hydroxides, carbon black, graphite, wood flour and flours or fibers of other natural products, synthetic fibers.

13. Other additives, for example, plasticisers, lubricants, emulsifiers, pigments, rheology additives, catalysts, flow-control agents, optical brighteners, flameproofing agents, antistatic agents and blowing agents.

The costabilizers are added, for example, in concentrations of from 0.01 to 10%, based on the overall weight of the organic material to be stabilized.

The fillers and reinforcing agents (item 12 in the list) , for example talc, calcium carbonate, mica or kaolin, are added to the polyolefin in concentrations, for example, of from 0.01 to 40%, based on the overall weight of the polyolefin to be stabilized.

The fillers and reinforcing agents (item 12 in the list) , for example metal hydroxides, especially aluminium hydroxide or magnesium hydroxide, are added to the polyolefin in concentrations, for example, of from 0.01 to 60%, based on the overall weight of the polyolefin to be stabilized.

Carbon black as filler is added to the polyolefin in concentrations, judiciously, of from 0.01 to 5%, based on the overall weight of the polyolefin to be stabilized.

Glass fibers as reinforcing agents are added to the polyolefin in concentrations, judiciously, of from 0.01 to 20%, based on the overall weight of the polyolefin to be stabilized.

Further preferred compositions comprise in addition to components (a) to (e) further additives as well, especially alkaline earth metal salts of higher fatty acids, for example calcium stearate.

As a conventional stabilizer combination for processing polymeric organic materials, for example polyolefins, to form corresponding mouldings, the combination of a phenolic antioxidant with a secondary antioxidant based on an organic phosphite or phosphonite is recommended. Depending on the particular substrate and process, however, many polyolefin processors are forced to operate processes in the high-temperature range above about 280°C. By virtue of the incorporation of a novel processing stabilizer mixture of components (b), (c), (d) and (e), which is especially suitable for high-temperature applications, in particular in the temperature range above 300°C, industrial materials and mouldings, based for instance on high-density polyethylene, for example pipes and their technical variants (fittings), can be produced at a greater rate and with fewer rejects. Another advantage of this stabilizer mixture is that it can be employed in very small amounts. This leads to a reduction in the overall concentration of antioxidant relative to conventional stabilizer mixtures. Thus

the use of a low concentration of a stabilizer of the benzofuran-2-one type [component (b)] allows a reduction in the overall concentration of stabilizer by about a third in polyolefins, for example, which simultaneously represents an economic advantage.

The incorporation of components (b), (c), (d) and (e), and further additives if desired, into the polymeric, organic material is carried out by known methods, for example prior to or during shaping or else by applying the dissolved or dispersed stabilizer mixture to the polymeric organic material, with or without subsequent evaporation of the solvent. The stabilizer mixture of components (b), (c), (d) and (e) with or without further additives, can also be added in the form of a masterbatch, which contains these components in a concentration, for example, of from 2.5 to 25% by weight, to the materials that are to be stabilized.

The stabilizer mixture of components (b), (c), (d) and (e), with or without further additives, can also be added before or during polymerization or prior to crosslinking.

The stabilizer mixture of components (b), (c), (d) and (e), with or without further additives, can be incorporated in pure form or encapsulated in waxes, oils or polymers into the organic material that is to be stabilized.

The stabilizer mixture of components (b), (c), (d) and (e), with or without further additives, can also be sprayed onto the polymer that is to be stabilized. It is able to dilute other additives (for example the conventional additives indicated above) or their melts so that they too can be sprayed together with these additives onto the polymer that is to be stabilized. Addition by spraying on during the deactivation of the polymerization catalysts is particularly advantageous, it being possible to carry out spraying using, for example, the steam used for deactivation.

In the case of spherically polymerized polyolefins it may, for example, be advantageous to apply the stabilizer mixture of components (b), (c), (d) and (e), with or without other additives, by spraying.

The materials stabilized in this way can be employed in a wide variety of forms, for example as films, fibres, tapes, moulding compositions, profiles or as binders for coating materials, especially powder coatings, adhesives or putties.



The polyolefins stabilized in this way can likewise be employed in a wide variety of forms, especially as thick-layer polyolefin mouldings which are in long-term contact with extractive media, , for example pipes for liquids or gases, films, geomembranes, strips, profiles or tanks.

The preferred thick-layer polyolefin mouldings have a layer thickness of from 1 to 50 mm, in particular from 1 to 30 mm, for example from 2 to 10 mm.

As already mentioned, the organic materials to be protected are preferably organic, especially synthetic, polymers. In this context, thermoplastic materials are protected with particular advantage. Attention should be drawn above all in this context to the outstanding activity of the novel stabilizer mixture of components (b), (c), (d) and (e) as in-process stabilizers (heat stabilizers). For this purpose they are advantageously added to the polymer prior to or during its processing. However, other polymers too (for example elastomers) or lubricants or hydraulic fluids can be stabilized against degradation, for example light-induced or thermo-oxidative degradation. Elastomers can be taken from the above listing of possible organic materials.

The lubricants and hydraulic fluids in question are based, for example, on mineral oils or synthetic oils or on mixtures thereof. The lubricants are familiar to the skilled worker and are described in the relevant technical literature, for example in Dieter Klamann, "Schmierstoffe und verwandte Produkte" (Verlag Chemie, Weinheim, 1982), in Schewe-Kobek, "Das Schmiermittel-Taschenbuch" (Dr. Alfred Hüthig-Verlag, Heidelberg, 1974) and in "Ullmanns Enzyklopädie der technischen Chemie", Vol.13, pages 85-94 (Verlag Chemie, Weinheim, 1977).

A preferred embodiment of the present invention is therefore the use of components (b), (c), (d) and (e) as stabilizers, especially in-process stabilizers (thermal stabilizers), for organic materials, especially thermoplastic polymers, against oxidative, thermal or light-induced degradation.

The present invention also relates to a stabilizer mixture comprising (i) at least one compound of the benzofuran-2-one type, (ii) at least one compound from the group of the orga-

nic phosphites or phosphonites, (iii) at least one compound from the group of the phenolic antioxidants, and (iv) at least one compound from the group of the sterically hindered amines.

Preference is also given to stabilizer mixtures in which the weight ratio of the components (i) : (ii) : (iii) : (iv) is from 10 : 1 : 1 : 0.1 to 0.01 : 1 : 10 : 100, in particular from 5 : 1 : 1 : 0.1 to 0.01 : 1 : 1 : 10.

The novel stabilizer mixture of components (b), (c), (d) and (e) features excellent stability to hydrolysis and advantageous colour behaviour, i.e. little discoloration of the organic material during processing.

Organic materials which are stabilized with the components of the present invention are particularly well protected against light-induced degradation.

The present invention also relates to a process for stabilizing an organic material against oxidative, thermal or light-induced degradation, which comprises incorporating in or applying to said material at least one each of components (b), (c), (d) and (e).

Preference is given to a process for stabilizing polyolefins that are in long-term contact with extractive media, wherein the polyolefins are thick-layer polyolefin mouldings and have a layer thickness of from 1 to 50 mm, in particular from 1 to 30 mm, for example from 2 to 10 mm, which comprises incorporating in or applying to said polyolefins at least one each of components (b), (c), (d) and (e).

Also of particular interest is a process for stabilizing thick-layer polyolefin mouldings that are in long-term contact with extractive media, wherein the thick-layer polyolefin mouldings are pipes or geomembranes, which comprises incorporating in or applying to said mouldings at least one each of components (b), (c), (d) and (e).

The term geomembranes refers to films which are employed, for example, in landfill sites and are required to have a service life of up to 300 years.

Extractive media are, for example, liquid or gaseous inorganic or organic materials.

Examples of gaseous inorganic materials are oxygen; nitrogen; oxides of nitrogen; for example NO, laughing gas or NO<sub>2</sub>; oxides of sulfur, for example sulfur dioxide; halogens, for example fluorine or chlorine; Brönstedt acids, for example hydrofluoric acid, hydrochloric acid, hydrobromic acid, hydroiodic acid or hydrocyanic acid; or bases, for example ammonia.

Examples of gaseous organic materials are C<sub>1</sub>-C<sub>4</sub>alkanes, for example methane, ethane, propane or butane; carbon monoxide; carbon dioxide; or phosgene.

Examples of liquid inorganic materials are water, chlorinated drinking water or aqueous salt solutions, for example sodium chloride solution (brine) or sodium sulfate solution; bromine; acid halides, e.g. titanium tetrachloride, thionyl chloride, nitrosyl chloride or trimethylsilyl chloride; alkalis, for example aqueous sodium hydroxide (NaOH), aqueous potassium hydroxide (KOH), aqueous ammonia solution, aqueous sodium bicarbonate solution or aqueous sodium carbonate solution.

Examples of liquid organic materials are organic solvents or liquid organic reagents.

Examples of organic solvents are aliphatic hydrocarbons, for example pentane, hexane, heptane, octane, petroleum spirit, nonane or decane; alcohols, for example methanol, ethanol, isopropanol, butanol, pentanol, amyl alcohol, cyclohexanol, pentaerythritol, ethylene glycol, ethylene diglycol, methylcellosolve, polyethylene glycol or glycerol; ketones, for example acetone, diethyl ketone, methyl ethyl ketone, diphenyl ketone or cyclohexanone; ethers, for example diethyl ether, dibutyl ether, tetrahydrofuran or dioxane; aromatic hydrocarbons, for example benzene, toluene or xylene; heterocyclic solvents, for example furan, pyridine, 2,6-lutidine or thiophene; dipolar aprotic solvents, for example dimethylformamide, diethylacetamide or acetonitrile; or surfactants.

For the purposes of the present invention, extractive media are also mixtures and solutions, especially aqueous mixtures, emulsions or solutions, of liquid or gaseous inorganic and organic materials as listed above.

Of particular interest are those extractive media which are important in the chemical industry or in landfill sites.

A preferred embodiment of the present invention is therefore also the use of a stabilizer mixture of components (b), (c), (d) and (e), with or without further additives, for improving the stability of polyolefins that are in long-term contact with extractive media.

The preferred components (b), (c), (d) and (e) for the use as stabilizers, the process for stabilizing and the stabilizer mixture are the same as those described for the compositions with an organic material.

The examples which follow illustrate the invention in more detail. Parts and percentages are by weight.

Example 1: Stabilizing polyethylene that is in long-term contact with water.

0.1% by weight of calcium stearate and the stabilizers listed in Table 1 are added dry to a polyethylene polymer (Hostalen® CRP 100; PE-HD) taken directly from the reactor, and the additives are incorporated in a Pappenmaier mixer (type 20) for two minutes (Examples 1a to 1c).

Table 1:

Examples	Stabilizers	Amount (% by wt.)
Example 1a <sup>a)</sup>	Irgafos®168 <sup>c)</sup> Irganox®1010 <sup>d)</sup> Chimassorb®944 <sup>e)</sup>	0.10 0.10 0.20
Example 1b <sup>b)</sup>	Verbindung (101) <sup>f)</sup> Irgafos®168 <sup>c)</sup> Irganox®1010 <sup>d)</sup> Chimassorb®944 <sup>e)</sup>	0.05 0.10 0.05 0.20
Example 1c <sup>b)</sup>	Verbindung (101) <sup>f)</sup> Irgafos®168 <sup>c)</sup> Irganox®1010 <sup>d)</sup> Chimassorb®119 <sup>g)</sup>	0.05 0.10 0.05 0.20

a) Comparison example.

- b) Example according to the invention.
- c) Irgafos®168 (Ciba Spezialitätenchemie AG) is tris(2,4-di-tert-butylphenyl) phosphite.
- d) Irganox®1010 (Ciba Spezialitätenchemie AG) is the pentaerythritol ester of 3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionic acid (compound of the formula VIc).
- e) Chimassorb®944 (Ciba Spezialitätenchemie AG) denotes linear or cyclic condensation products prepared from N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine and 4-tert-octylamino-2,6-dichlor-1,3,5-triazine [compound of the formula (H5)].
- f) Compound (101) is a mixture of about 85 parts by weight of the compound of the formula Va and about 15 parts by weight of the compound of the formula Vb.
- g) Chimassorb®119 (Ciba Spezialitätenchemie AG) denotes condensation products prepared from 2-chloro-4,6-di-(4-n-butylamino-1,2,2,6,6-pentamethylpiperidyl)-1,3,5-triazine and 1,2-bis(3-aminopropylamino)ethane [compound of the formula (H6)].

Irgafos®168, Irganox®1010, Chimassorb®944 and Chimassorb®119 are protected trade names of the company Ciba Spezialitätenchemie AG

In a Schwabenthan extruder the stabilized polyethylene is homogenized at 220°C and is processed at 300°C to granules. For the extraction experiments in water, test sheets measuring 44 mm by 20 mm by 2 mm are pressed from the granules of the individual formulations (Examples 1a to 1c) using a bench press. To facilitate the demoulding of the test sheets, pressing is carried out between two aluminium foils.

The stabilizer extraction experiments were carried out with deionized water. The thermal conditioning of the extraction containers takes place in a convection oven from Heraeus (Hanau, Germany) with a maximum temperature deviation of 1.5°C. For the extraction experiments below the boiling point of water, glass vessels are used. In the case of a water temperature of 105°C, stainless steel pressure vessels are employed. Owing to the risk of oversaturation of the water with stabilizer, the amount of liquid for the experiments is fixed at about 400 ml for about 70 g of polymer and the water is replaced at regular intervals of time, specifically after each sampling, by fresh water.

The test sheets are exposed at 80°C for 50 days to the experimental conditions described above. After the end of the extraction experiments, the residual stabilizer content and oxidation induction time (OIT) of the test sheets are measured.

The residual content of the sterically hindered phenol, Irganox®1010, is determined with the aid of an internal standard in a HPLC instrument of the type Spectra Physics SP 8800 with autosampler and UV/VIS detector of the type Spectra 200. The chromatography is carried out at room temperature with a column of the hyperchrome type 125 x 4.6 mm, packed with Nucleosil C 185 µm. The injection volume is 14 µl at a flow rate of 1.5 ml/minute. UV detection takes place at 270 nm.

The residual content of the triazine-containing sterically hindered amines, Chimassorb® 944 and Chimassorb®119, is determined with a UV spectrometer of the Perkin Elmer Lambda 15 type by measuring the difference between the absorbances at 246.4 and 300 nm.

The oxidation induction time is determined by means of a "DuPont instrument 910 differential scanning calorimeter" from TA Instruments (Alzenau, Germany) and with a sample amount of from 5 to 10 mg, and describes the period of time, in minutes, under constant thermal stress (190°C/O<sub>2</sub>) until complete decomposition of the polyethylene sample begins. The longer the oxidation induction time, the better the stabilization of polyethylene and the more stable the polyethylene with respect to extracted water which is in long-term contact with the polyethylene.

The results show that the stability of polyolefins that are in long-term contact with extracted media is improved if the stabilizer mixture comprises a novel component (b), (c), (d) and (e). The results are summarized in Table 2.

Table 2: Water storage for 50 days at 80°C

Example	Stabilizer	Residual stabilizer content after 50 days at 80°C	Oxidation induction time in minutes
1a <sup>a)</sup>	0,10% Irgafos®168 <sup>c)</sup> 0,10% Irganox®1010 <sup>d)</sup> 0,20% Chimassorb®944 <sup>e)</sup>	52% 57%	34
1b <sup>b)</sup>	0,05% Compound (101) <sup>f)</sup> 0,10% Irgafos®168 <sup>c)</sup> 0,05% Irganox®1010 <sup>d)</sup> 0,20% Chimassorb®944 <sup>e)</sup>	56% 71%	38
1c <sup>b)</sup>	0,05% Compound (101) <sup>f)</sup> 0,10% Irgafos®168 <sup>c)</sup> 0,05% Irganox®1010 <sup>d)</sup> 0,20% Chimassorb®944 <sup>e)</sup>	57% 70%	39

Footnotes a) to g) see Table 1.

**Example 2:** Stabilizing polypropylene in the case of multiple extrusion and at especially high temperatures.

1.5 kg of polypropylene powder (Profax®6501), which has been initially stabilized with 0.008% of Irganox®1076 (compound of the formula VIb) (with a melt index of 3.2 measured at 230°C and under 2.16 kg), are mixed with 0.10% of calcium stearate and 0.015 to 0.20% of the stabilizers listed in Table 3. This mixture is extruded in an extruder having a barrel diameter of 20 mm and a length of 400 mm at 100 revolutions per minute, the maximum extruder temperature being set at 280, 300, 320 and 340°C. For cooling, the extrudate is drawn through a water bath and then granulated. These granules are extruded repeatedly. After 5 extrusions, the melt index is measured (at 230°C under 2.16 kg). A large increase in the melt index denotes severe chain breakdown and hence poor stabilization. The results are summarized in Table 3. They show that the stability of polypropylene is improved if the stabilizer mixture comprises a novel component (b), (c), (d) and (e).

Table 3:

Example	Stabilizers	Amount (% by wt.)	Melt index after 5 extrusions
Example 2a <sup>a)</sup>	Irgafos <sup>®</sup> 168 <sup>c)</sup> Irganox <sup>®</sup> 1010 <sup>d)</sup> Chimassorb <sup>®</sup> 944 <sup>e)</sup>	0.10 0.10 0.20	17.5
Example 2b <sup>b)</sup>	Compound (101) <sup>f)</sup> Irgafos <sup>®</sup> 168 <sup>c)</sup> Irganox <sup>®</sup> 1010 <sup>d)</sup> Chimassorb <sup>®</sup> 944 <sup>e)</sup>	0.015 0.10 0.05 0.10	8.6
Example 2c <sup>b)</sup>	Compound (101) <sup>f)</sup> Irgafos <sup>®</sup> 168 <sup>c)</sup> Irganox <sup>®</sup> 1010 <sup>d)</sup> Chimassorb <sup>®</sup> 119 <sup>g)</sup>	0.015 0.10 0.05 0.10	8.0

Footnotes a) to g) see Table 1 of Example 1.

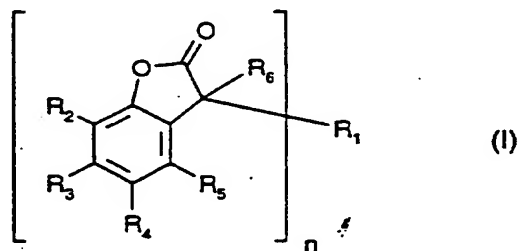


WHAT IS CLAIMED IS:

1. A composition comprising

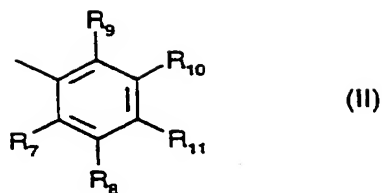
- a) an organic material which is subject to oxidative, thermal or light-induced degradation,
- b) at least one compound of the benzofuran-2-one type,
- c) at least one compound from the group of the organic phosphites or phosphonites,
- d) at least one compound from the group of the phenolic antioxidants, and
- e) at least one compound from the group of the sterically hindered amines.

2. A composition according to claim 1, comprising as component (b) a compound of the formula I



in which, if n is 1,

R<sub>1</sub> is unsubstituted or C<sub>1</sub>-C<sub>4</sub>alkyl-, C<sub>1</sub>-C<sub>4</sub>alkoxy-, C<sub>1</sub>-C<sub>4</sub>alkylthio-, hydroxyl-, halo-, amino-, C<sub>1</sub>-C<sub>4</sub>alkylamino-, phenylamino- or di(C<sub>1</sub>-C<sub>4</sub>alkyl)amino-substituted naphthyl, phenanthryl, anthryl, 5,6,7,8-tetrahydro-2-naphthyl, 5,6,7,8-tetrahydro-1-naphthyl, thienyl, benzo[b]thienyl, naphtho[2,3-b]thienyl, thianthrenyl, dibenzofuryl, chromenyl, xanthenyl, phenoxathiinyl, pyrrolyl, imidazolyl, pyrazolyl, pyrazinyl, pyrimidinyl, pyridazinyl, indoliziny, isoindolyl, indolyl, indazolyl, purinyl, quinoliziny, isoquinolyl, quinolyl, phthalazinyl, naphthyridinyl, quinoxalinyl, quinazolinyl, cinnolinyl, pteridinyl, carbazolyl, β-carbolinyl, phenanthridinyl, acridinyl, perimidinyl, phenanthrolinyl, phenazinyl, isothiazolyl, phenothiazinyl, isoxazolyl, furazanyl, biphenyl, terphenyl, fluorenyl or phenoxazinyl, or R<sub>1</sub> is a radical of the formula II



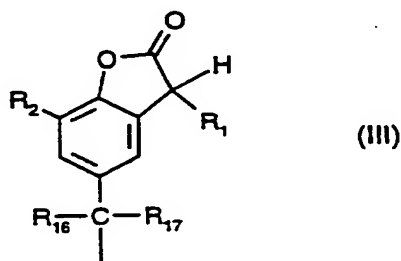
and

if n is 2,

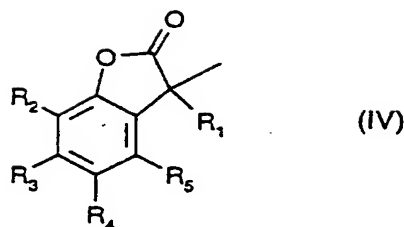
$R_1$  is unsubstituted or  $C_1$ - $C_4$ alkyl- or hydroxyl-substituted phenylene or naphthylene; or is  $-R_{12}-X-R_{13}-$ ,

$R_2$ ,  $R_3$ ,  $R_4$  and  $R_5$  independently of one another are hydrogen, chlorine, hydroxyl,  $C_1$ - $C_{25}$ alkyl,  $C_7$ - $C_9$ phenylalkyl, unsubstituted or  $C_1$ - $C_4$ alkyl-substituted phenyl; unsubstituted or  $C_1$ - $C_4$ alkyl-substituted  $C_5$ - $C_8$ cycloalkyl;  $C_1$ - $C_{18}$ alkoxy,  $C_1$ - $C_{18}$ alkylthio,  $C_1$ - $C_4$ alkylamino, di( $C_1$ - $C_4$ alkyl)-amino,  $C_1$ - $C_{25}$ alkanoyloxy,  $C_1$ - $C_{25}$ alkanoylamino,  $C_3$ - $C_{25}$ alkenoyloxy,  $C_3$ - $C_{25}$ alkanoyloxy which is interrupted by oxygen, sulfur or  $\text{>N}-R_{14}$ ;  $C_6$ - $C_9$ cycloalkylcarbonyloxy, benzoyloxy or

$C_1$ - $C_{12}$ alkyl-substituted benzoyloxy; or else the radicals  $R_2$  and  $R_3$  or the radicals  $R_3$  and  $R_4$  or the radicals  $R_4$  and  $R_5$ , together with the carbon atoms to which they are attached, form a benzo ring,  $R_4$  is additionally  $-(CH_2)_p-COR_{15}$  or  $-(CH_2)_qOH$  or, if  $R_3$ ,  $R_5$  and  $R_6$  are hydrogen,  $R_4$  is additionally a radical of the formula III



in which  $R_1$  is defined as indicated above for  $n = 1$ ,  
 $R_6$  is hydrogen or a radical of the formula IV



where  $R_4$  is not a radical of the formula III and  $R_1$  is defined as indicated above for  $n = 1$ ,  $R_7$ ,  $R_8$ ,  $R_9$ ,  $R_{10}$  and  $R_{11}$  independently of one another are hydrogen, halogen, hydroxyl,

$C_1$ - $C_{25}$ alkyl,  $C_2$ - $C_{25}$ alkyl interrupted by oxygen, sulfur or  $\text{>N}-R_{14}$  ;  $C_1$ - $C_{25}$ alkoxy,

$C_2$ - $C_{25}$ alkoxy interrupted by oxygen, sulfur or  $\text{>N}-R_{14}$  ;  $C_1$ - $C_{25}$ alkylthio,  $C_3$ - $C_{25}$ alkenyl,

$C_3$ - $C_{25}$ alkenyloxy,  $C_3$ - $C_{25}$ alkynyl,  $C_3$ - $C_{25}$ alkynyloxy,  $C_7$ - $C_9$ phenylalkyl,  $C_7$ - $C_9$ phenylalkoxy, unsubstituted or  $C_1$ - $C_4$ alkyl-substituted phenyl; unsubstituted or  $C_1$ - $C_4$ alkyl-substituted phenoxy; unsubstituted or  $C_1$ - $C_4$ alkyl-substituted  $C_5$ - $C_8$ cycloalkyl; unsubstituted or  $C_1$ - $C_4$ alkyl-substituted  $C_5$ - $C_8$ cycloalkoxy;  $C_1$ - $C_4$ alkylamino, di( $C_1$ - $C_4$ alkyl)amino,  $C_1$ - $C_{25}$ alkanoyl,

$C_3$ - $C_{25}$ alkanoyl interrupted by oxygen, sulfur or  $\text{>N}-R_{14}$  ;  $C_1$ - $C_{25}$ alkanoyloxy,  $C_3$ - $C_{25}$ alka-

noyloxy interrupted by oxygen, sulfur or  $\text{>N}-R_{14}$  ;  $C_1$ - $C_{25}$ alkanoylamino,  $C_3$ - $C_{25}$ alkenoyl,

$C_3$ - $C_{25}$ alkenoyl interrupted by oxygen, sulfur or  $\text{>N}-R_{14}$  ;  $C_3$ - $C_{25}$ alkenoyloxy,  $C_3$ - $C_{25}$ alke-

noyloxy interrupted by oxygen, sulfur or  $\text{>N}-R_{14}$  ;  $C_6$ - $C_9$ cycloalkylcarbonyl,  $C_6$ - $C_9$ cycloalkylcarbonyloxy, benzoyl or  $C_1$ - $C_{12}$ alkyl-substituted benzoyl; benzoyloxy or  $C_1$ - $C_{12}$ alkyl-sub-

stituted benzoyloxy;  $\text{—O—}\overset{\overset{R_{18}}{|}}{\underset{\underset{R_{19}}{|}}{C}}\text{—}\overset{\overset{O}{||}}{C}\text{—}R_{15}$  or  $\text{—O—}\overset{\overset{R_{20}}{|}}{\underset{\underset{H}{|}}{C}}\text{—}\overset{\overset{R_{21}}{|}}{\underset{\underset{R_{22}}{|}}{C}}\text{—O—}R_{23}$  , or else, in formu-

la II, the radicals  $R_7$  and  $R_8$  or the radicals  $R_8$  and  $R_{11}$ , together with the carbon atoms to which they are attached, form a benzo ring,

$R_{12}$  and  $R_{13}$  independently of one another are unsubstituted or  $C_1$ - $C_4$ alkyl-substituted phenylene or naphthylene,

$R_{14}$  is hydrogen or  $C_1$ - $C_8$ alkyl,

$R_{15}$  is hydroxyl,  $\left[ -O^- \frac{1}{r} M^{r+} \right]$ ,  $C_1$ - $C_{18}$ alkoxy or  $-N \begin{matrix} R_{24} \\ R_{25} \end{matrix}$ ,

$R_{16}$  and  $R_{17}$  independently of one another are hydrogen,  $CF_3$ ,  $C_1$ - $C_{12}$ alkyl or phenyl, or  $R_{16}$  and  $R_{17}$ , together with the C atom to which they are attached, form a  $C_5$ - $C_8$ cycloalkylidene ring which is unsubstituted or substituted from 1 to 3 times by  $C_1$ - $C_4$ alkyl;

$R_{18}$  and  $R_{19}$  independently of one another are hydrogen,  $C_1$ - $C_4$ alkyl or phenyl,

$R_{20}$  is hydrogen or  $C_1$ - $C_4$ alkyl,

$R_{21}$  is hydrogen, unsubstituted or  $C_1$ - $C_4$ alkyl-substituted phenyl;  $C_1$ - $C_{25}$ alkyl,  $C_2$ - $C_{25}$ alkyl

interrupted by oxygen, sulfur or  $\text{>N}-R_{14}$ ;  $C_7$ - $C_9$ phenylalkyl which is unsubstituted or

substituted on the phenyl radical from 1 to 3 times by  $C_1$ - $C_4$ alkyl;  $C_7$ - $C_{25}$ phenylalkyl which is unsubstituted or substituted on the phenyl radical from 1 to 3 times by  $C_1$ - $C_4$ alkyl and

interrupted by oxygen, sulfur or  $\text{>N}-R_{14}$ , or else the radicals  $R_{20}$  and  $R_{21}$ , together with

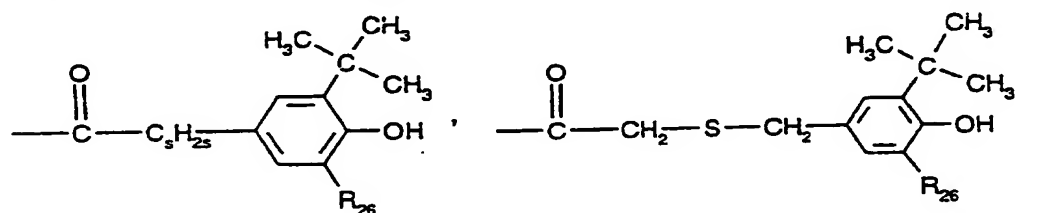
the carbon atoms to which they are attached, form a  $C_5$ - $C_{12}$ cycloalkylene ring which is unsubstituted or substituted from 1 to 3 times by  $C_1$ - $C_4$ alkyl;

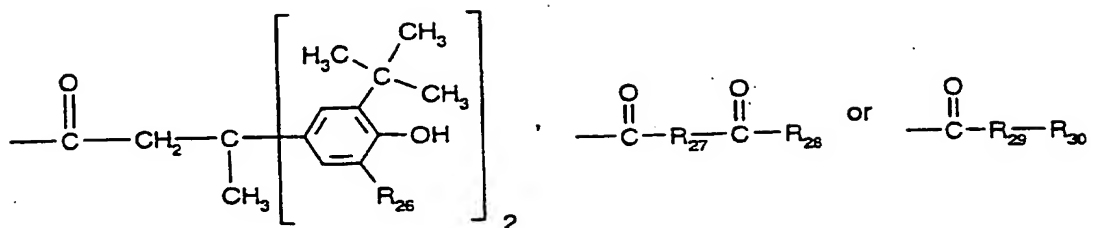
$R_{22}$  is hydrogen or  $C_1$ - $C_4$ alkyl,

$R_{23}$  is hydrogen,  $C_1$ - $C_{25}$ alkanoyl,  $C_3$ - $C_{25}$ alkenoyl,  $C_3$ - $C_{25}$ alkanoyl interrupted by oxygen, sulfur

or  $\text{>N}-R_{14}$ ;  $C_2$ - $C_{25}$ alkanoyl substituted by a di( $C_1$ - $C_6$ alkyl)phosphonate group;

$C_6$ - $C_9$ cycloalkylcarbonyl, thenoyl, furoyl, benzoyl or  $C_1$ - $C_{12}$ alkyl-substituted benzoyl;

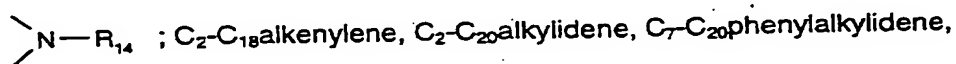




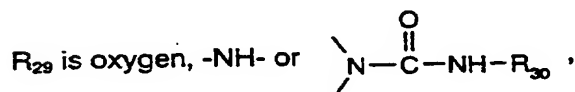
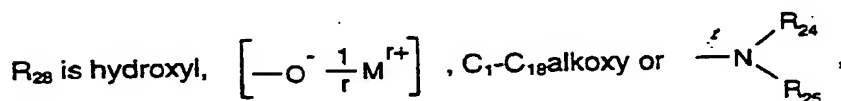
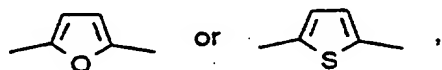
R<sub>24</sub> and R<sub>25</sub> independently of one another are hydrogen or C<sub>1</sub>-C<sub>18</sub>alkyl,

R<sub>26</sub> is hydrogen or C<sub>1</sub>-C<sub>8</sub>alkyl,

R<sub>27</sub> is a direct bond, C<sub>1</sub>-C<sub>18</sub>alkylene, C<sub>2</sub>-C<sub>18</sub>alkylene interrupted by oxygen, sulfur or



C<sub>5</sub>-C<sub>8</sub>cycloalkylene, C<sub>7</sub>-C<sub>8</sub>bicycloalkylene, unsubstituted or C<sub>1</sub>-C<sub>4</sub>alkyl-substituted phenylene,



R<sub>30</sub> is C<sub>1</sub>-C<sub>18</sub>alkyl or phenyl,

R<sub>31</sub> is hydrogen or C<sub>1</sub>-C<sub>18</sub>alkyl,

M is an r-valent metal cation,

X is a direct bond, oxygen, sulfur or -NR<sub>31</sub>-,

n is 1 or 2,

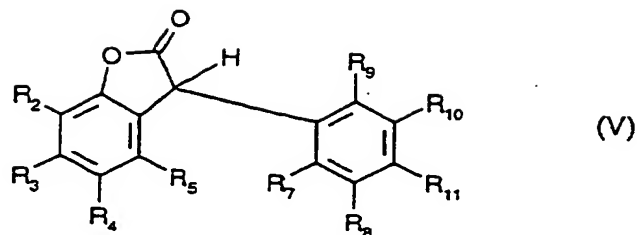
p is 0, 1 or 2,

q is 1, 2, 3, 4, 5 or 6,

r is 1, 2 or 3, and

s is 0, 1 or 2.

3. A composition according to claim 1, comprising as component (b) a compound of the formula V



in which

$R_2$  is hydrogen or  $C_1$ - $C_6$ alkyl,

$R_3$  is hydrogen,

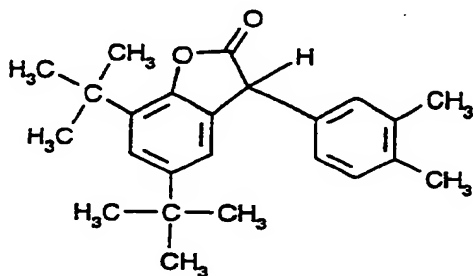
$R_4$  is hydrogen or  $C_1$ - $C_6$ alkyl,

$R_5$  is hydrogen,

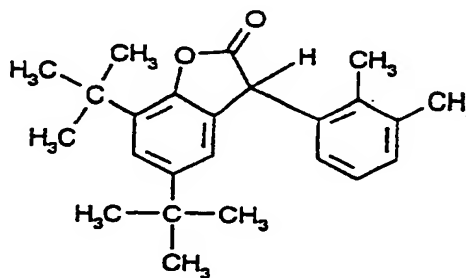
$R_7$ ,  $R_8$ ,  $R_9$ ,  $R_{10}$  and  $R_{11}$  independently of one another are hydrogen,  $C_1$ - $C_4$ alkyl or  $C_1$ - $C_4$ alkoxy, with the proviso that at least two of the radicals  $R_7$ ,  $R_8$ ,  $R_9$ ,  $R_{10}$  or  $R_{11}$  are hydrogen.

?

4. A composition according to claim 1, in which component (b) is a compound of the formula Va or Vb



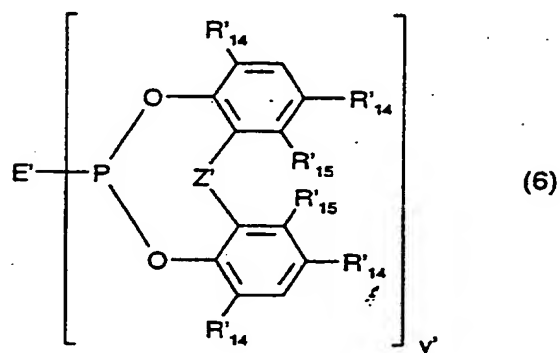
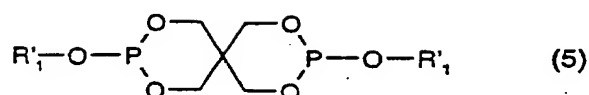
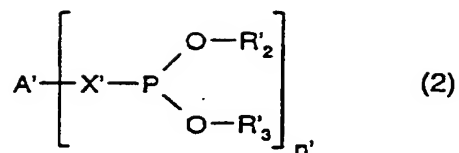
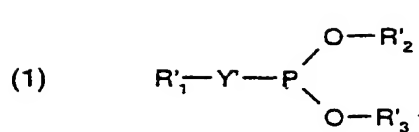
(Va)



(Vb)

or a mixture of the two compounds of the formula Va and Vb.

5. A composition according to claim 1, comprising as component (c) a compound of the formula 1, 2, 5 or 6



in which

$n'$  is the number 2 and  $y'$  is the number 1, 2 or 3;

$A'$  is  $C_2$ - $C_{18}$ alkylene, p-phenylene or p-biphenylene,

$E'$ , if  $y'$  is 1, is  $C_1$ - $C_{18}$ alkyl,  $-OR'_1$  or fluorine;

$E'$ , if  $y'$  is 2, is p-biphenylene,

$E'$ , if  $y'$  is 3, is  $N(CH_2CH_2O)_3$ ,

$R'_1$ ,  $R'_2$  and  $R'_3$  independently of one another are  $C_1$ - $C_{18}$ alkyl,  $C_7$ - $C_9$ phenylalkyl, cyclohexyl, phenyl, or phenyl substituted by 1 to 3 alkyl radicals having in total 1 to 18 carbon atoms;

$R'_{14}$  is hydrogen or  $C_1$ - $C_9$ alkyl,

$R'_{15}$  is hydrogen or methyl;

$X'$  is a direct bond,

$Y'$  is oxygen,

$Z'$  is a direct bond or  $-\text{CH}(R'_{16})-$ , and

$R'_{16}$  is  $C_1$ - $C_4$ alkyl.



**R<sub>1</sub> is C<sub>1</sub>-C<sub>4</sub>alkyl,**

**X is methylene,**

oxygen

X is  $\text{---CH}_2\text{---CH}_2\text{---}\overset{\overset{\text{O}}{\parallel}}{\text{C}}\text{---Y---}$

$$Y \text{ is } \text{---CH---CH}_2\text{---}\overset{\overset{\text{||}}{\text{O}}}{\text{C}}\text{---Y---}$$

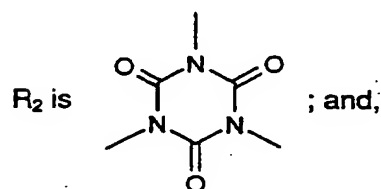
where Y is attached to R<sub>2</sub>, and



$R_2$  is  $C_2$ - $C_{12}$ alkylene,  $C_4$ - $C_{12}$ alkylene interrupted by oxygen or sulfur; or, if Y is -NH-,  $R_2$  is additionally a direct bond; and,

if n is 3,

X is methylene or  $\text{---CH}_2\text{---}\overset{\text{O}}{\parallel}\text{C---O---CH}_2\text{---CH}_2\text{---}$ , where the ethylene group is attached to  $R_2$ , and



if n is 4,

X is  $\text{---CH}_2\text{---CH}_2\text{---}\overset{\text{O}}{\parallel}\text{C---Y---}$ , where Y is attached to  $R_2$ , and  
 $R_2$  is  $C_4$ - $C_{10}$ alkanetetrayl.

8. Composition according to claim 7, in which in the compound of the formula VI, if n is 1,  $R_2$  is  $C_1$ - $C_{20}$ alkyl.

9. A composition according to claim 7, in which in the compound of the formula VI,

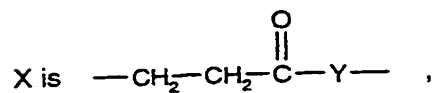
if n is 2,

$R_2$  is  $C_2$ - $C_8$ alkylene,  $C_4$ - $C_8$ alkylene interrupted by oxygen or sulfur; or, if Y is -NH-,  $R_2$  is additionally a direct bond; and,

if n is 4,

$R_2$  is  $C_4$ - $C_8$ alkanetetrayl.

10. A composition according to claim 7, in which in the compound of the formula VI  
 $R_1$  is methyl or tert-butyl,  
 $n$  is 1, 2 or 4,



Y is oxygen or -NH-; and,

if  $n$  is 1,

$R_2$  is  $C_{14}$ - $C_{18}$ alkyl; and,

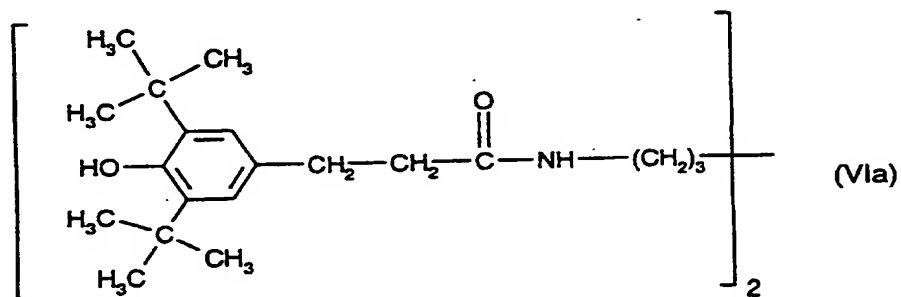
if  $n$  is 2,

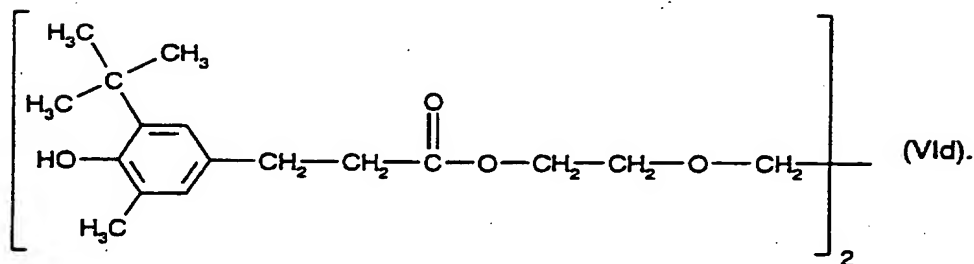
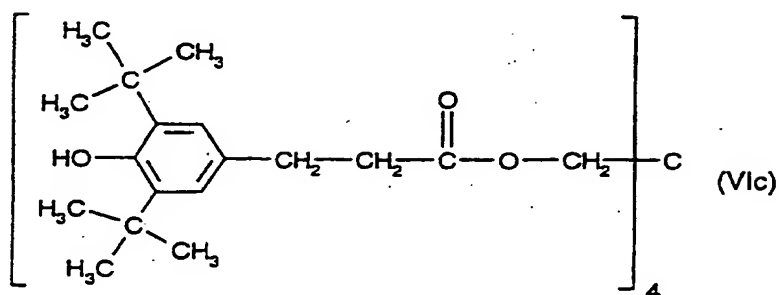
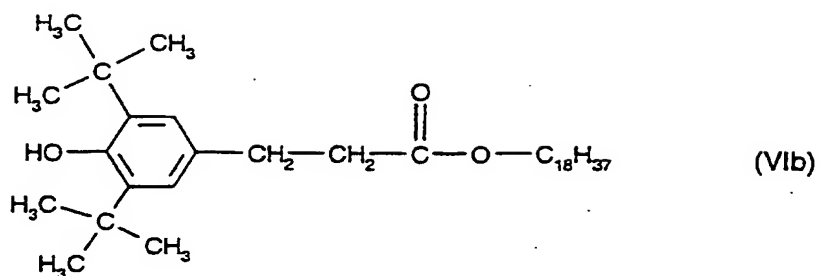
$R_2$  is  $C_4$ - $C_6$ alkylene, or is  $C_4$ - $C_6$ alkylene interrupted by oxygen; and,

if  $n$  is 4,

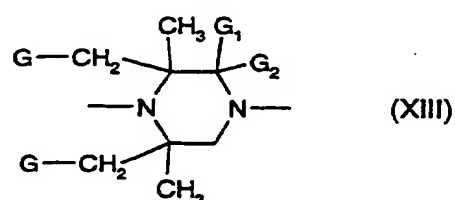
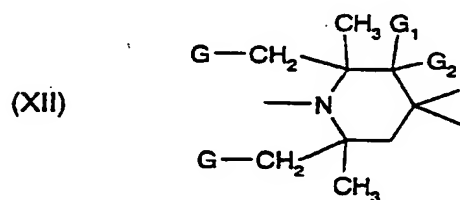
$R_2$  is  $C_4$ - $C_6$ alkanetetrayl.

11. A composition according to claim 1, in which component (d) is at least one compound of the formula VIa, VIb, VIc or VId





12. A composition according to claim 1, in which component (e) comprises at least one radical of the formula XII or XIII

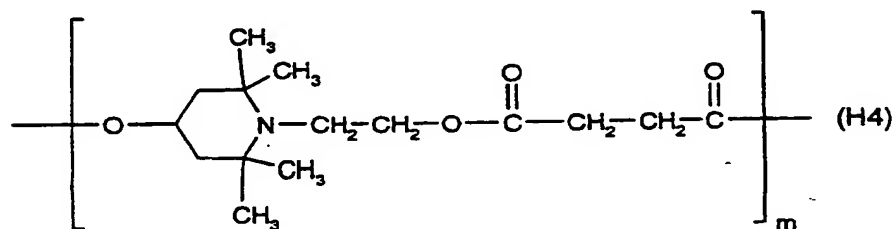
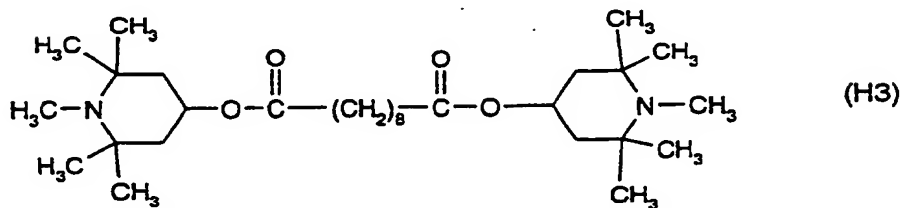
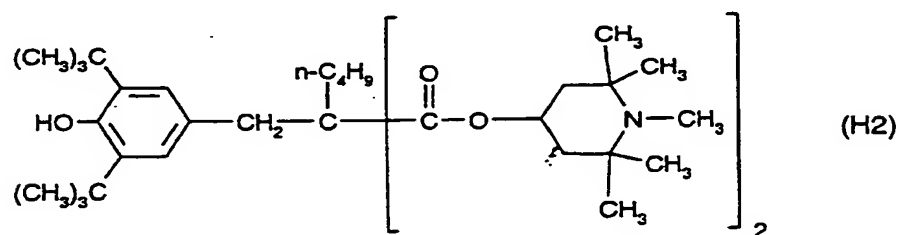
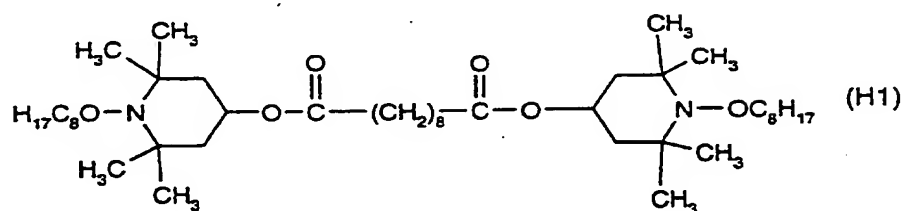


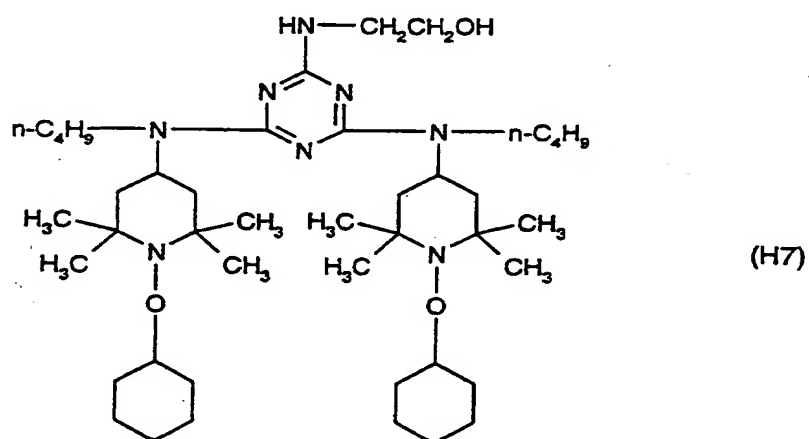
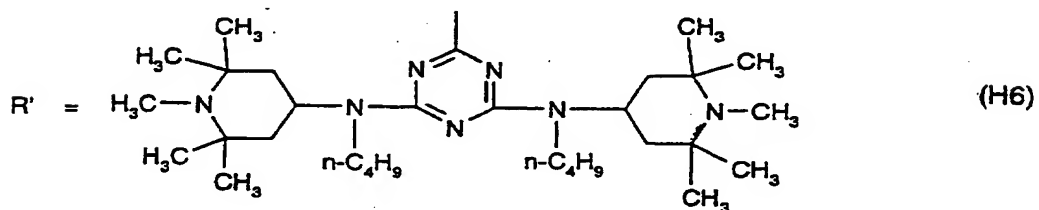
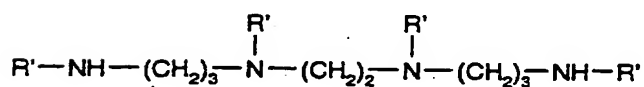
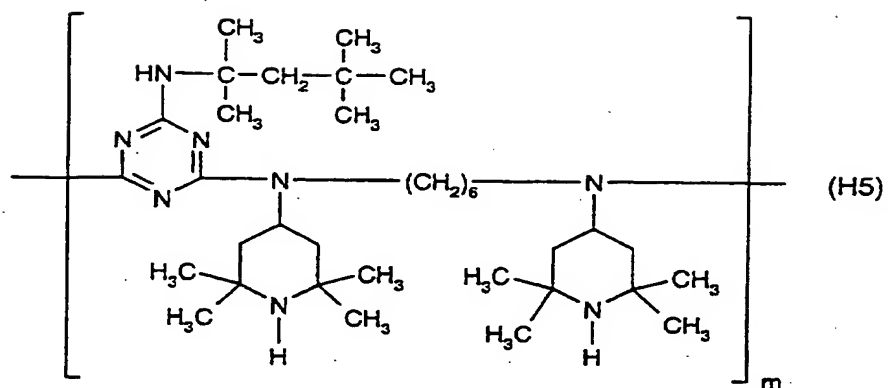
in which

G is hydrogen or methyl, and

G<sub>1</sub> and G<sub>2</sub> are hydrogen, methyl or together are oxygen.

13. A composition according to claim 1, in which component (e) is a compound of the formula H1, H2, H3, H4, H5, H6 or H7





in which

**m is a number from the range from 2 to 200.**

14. A composition according to claim 1, comprising as component (a) a natural, semisynthetic or synthetic polymer.
15. A composition according to claim 1, comprising as component (a) a thermoplastic polymer.
16. A composition according to claim 1, comprising as component (a) a polyolefin.
17. A composition according to claim 1, comprising as component (a) polyethylene or polypropylene or a copolymer thereof with mono- and diolefins.
18. A composition according to claim 1, comprising as component (a) a thick-layer polyolefin moulding.
19. A composition according to claim 18, in which the thick-layer polyolefin moulding has a layer thickness of from 1 to 50 mm.
20. A composition according to claim 18, in which the thick-layer polyolefin moulding comprises polyolefin pipes or polyolefin geomembranes.
21. A composition according to claim 1, comprising as component (a) a polyolefin which is in long-term contact with extractive media.
22. A composition according to claim 21, in which the extractive medium is a liquid or gaseous inorganic or organic material.
23. A composition according to claim 1, in which component (b) is present in an amount of from 0.0005 to 5% based on the weight of component (a).
24. A composition according to claim 1, in which components (c), (d) and (e) are present in an amount of from 0.01 to 10% based on the weight of component (a).
25. A composition according to claim 1, comprising in addition to components (a), (b), (c), (d) and (e) further additives as well.

26. A stabilizer mixture comprising

- i) at least one compound of the benzofuran-2-one type,
- ii) at least one compound from the group of the organic phosphites or phosphonites,
- iii) at least one compound from the group of the phenolic antioxidants, and
- iv) at least one compound from the group of the sterically hindered amines.

27. A stabilizer mixture according to claim 26, in which the weight ratio of the components (i) : (ii) : (iii) : (iv) is from 10 : 1 : 1 : 0.1 to 0.01 : 1 : 10 : 100.

28. A process for stabilizing an organic material against oxidative, thermal or light-induced degradation, which comprises incorporating in or applying to said material at least one each of components (b), (c), (d) and (e) according to claim 1.

29. The use of a mixture of components (b), (c), (d) and (e) according to claim 1 as stabilizers for organic materials against oxidative, thermal or light-induced degradation.

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The  
Patent  
Office

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Application No: GB 9802277.5  
Claims searched: 1 to 29

Examiner: Miss M. M. Kelman  
Date of search: 14 April 1998

**Patents Act 1977**  
**Search Report under Section 17**

**Databases searched:**

UK Patent Office collections, including GB, EP, WO & US patent specifications, in:

UK Cl (Ed.P): C3K KCZ, C3M MA

Int Cl (Ed.6): C08K 5/00, 5/15, 5/3435

Other: ONLINE: CHABS, CLAIMS, JAPIO, RAPRA, WPI

**Documents considered to be relevant:**

Category	Identity of document and relevant passage		Relevant to claims
X,P	GB 2315070 A	CIBA SPECIALTY CHEMICALS see the claims, page 64, lines 11 to 19, page 65, lines 11 to 18, page 74, lines 21 to 22, and Examples 1,2,4 and 5	1 to 17,21,22,23,24,25,26,28,29
A,P	GB 2311528 A	CIBA SPECIALTY CHEMICALS	
A	GB 2293827 A	SANDOZ	
A	US 5516920 A	CIBA-GEIGY	

X Document indicating lack of novelty or inventive step  
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